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*Frankenstein: A monstrous Romanticism*

Königkrämer, Lobke

KNGLOB001

A minor dissertation submitted in partial fulfillment of the requirements for the award of the  
degree of Master of Arts in English Literature

Faculty of the Humanities

University of Cape Town

2010

COMPULSORY DECLARATION

This work has not been previously submitted in whole, or in part, for the award of any degree. It is my own work. Each significant contribution to, and quotation in, this dissertation from the work, or works, of other people has been attributed, and has been cited and referenced.

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## **Abstract**

The purpose of this thesis is to examine the relationship between Mary Shelley's first novel *Frankenstein* and her own understanding of Romanticism. The overarching theme is to illustrate how Mary Shelley navigates her criticism of Romanticism through the medium of Victor Frankenstein as a character. With the inspection of Victor Frankenstein some autobiographical similarities are drawn between the protagonist and Percy Bysshe Shelley. Another aim and extension of this autobiographical project is to examine how Percy Shelley's editing of the original manuscript of *Frankenstein* added or detracted from the plot. Finally, the genre implications of *Frankenstein* are examined in this thesis.

In the first chapter, Romanticism is examined in relation to how the Romantics themselves envisioned their ideology so as to ascertain which aspects Mary Shelley draws particular attention to. The Romantic theorists used in this section specifically, Abercrombie and Schueller, are used to highlight the fact that Romanticism can be defined as a unified system of belief. Certain tenets of this ideology are then shown to be the main points that Mary Shelley criticises.

In the second chapter, the autobiographical element of Mary Shelley's relationship with Percy Shelley is examined. The parallels between Victor Frankenstein and Percy Shelley are made apparent through the use of biographers Hoobler and Seymour. From that, the precise changes that Percy Shelley made to the original manuscript of *Frankenstein* are scrutinised with Mellor's insightful explication of the original that exists in the Bodleian Library. The conclusion of this chapter solidifies the argument of the first chapter, and as close attention is paid throughout both chapters to the novel as a primary source of confirmation, the complex navigations and articulations of Romanticism throughout *Frankenstein* are made apparent.

In the third chapter, attention is given specifically to the genre implications of *Frankenstein*, and the relationship and consistent oscillation between Romanticism and the Gothic is traced. The theorists used in this part of the thesis vary widely and include Botting, Golinski and Alwes. It is argued that in her destabilisation of Romanticism, Mary Shelley invariably incorporates the Gothic into her text. It is this complex weaving of genres which is particularly interesting in

relation to how Mary Shelley's disillusionment with Romanticism produces a text that has such a vast array of genre possibilities.

Finally, this thesis looks at the negative interpretation of Romanticism specifically in relation to Mary Shelley's critical expressions of its ideology in *Frankenstein*. As a cautionary tale, the consequences of Romantic principles unchecked by a societal conscience, Mary Shelley seems to have used *Frankenstein* as a way of expressing her disillusionment. The repercussions of what ultimately is an original story of a scientist who unleashes his creation without concern for its welfare are still present in the common consciousness of modern society.

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## Introduction

What terrified me will terrify others; and I need only describe the spectre which had haunted my midnight pillow. ~ Mary Shelley, *Author's Introduction to Frankenstein*, 1831 edition, (page 9).

When one examines Mary Shelley's *Frankenstein*, one is immediately struck by the remarkable complexity of it. Many critics assume that this complexity is due to Mary Shelley being who she was, namely the sole child of the politico social figures of William Godwin and Mary Wollstonecraft. This assumption is only intensified or elaborated by her later life, as her elopement with Percy Bysshe Shelley is seen as yet another connection to a brilliant mind. So often, her originality is read in some way or another reactionary to or induced by the people that surrounded her. This thesis sets out to argue that Mary Shelley was writing against the ideology of those that surrounded her, broadly categorised as Romanticism. This maintains in a sense the element against which she is reacting, but allows Mary Shelley insight and a scepticism that denies the passive nature attributed to her writing of *Frankenstein* merely because she was related to literary giants.

It is because of this that the theorists of Romanticism used in this thesis were chosen particularly for their unified vision of Romanticism. It is important to focus on theorists who do not deconstruct Romanticism, but rather, for the purpose of this argument, to illustrate how the Romantics saw their ideology as a unified system of belief. It is for this reason that the main articulation of what Romanticism can be defined as is taken jointly from Abercrombie and Schueller<sup>1</sup>. Although they are both not the most contemporary theorists, and their view of Romanticism is definitely not to be taken at the exclusion of other more modern interpretations, the fact is that they both try to establish a unified vision of Romantic ideology. Contemporary scholarship tends to either glorify Romanticism with a sense of nostalgia, as is demonstrated by Ferguson's understanding of *Frankenstein* as a positive affirmation of Romanticism (Ferguson 1992); or, on the opposite side of the debate, tends to fragment Romanticism and be stuck with trying to define but also not to define it as a complete entity. Even though there is validity to some of this type of wrangling, the aim of this thesis is not to become involved with how

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<sup>1</sup> Taken jointly from Abercrombie, Lascelles. *Romanticism*. Londond: Martine Secker (Ltd), 1927. and Schueller, Herbert M. "Romanticism Reconsidered." *The Journal of Aesthetics and Art Criticism*, 1962:359 - 368.

Romanticism should be defined by modern scholarship, but more pointedly, how Romanticism is viewed and utilised by Mary Shelley within *Frankenstein* to destabilise and warn against Romantic idealism. Therefore the focus of this thesis is particularly on Mary Shelley's views of Romanticism as they are expressed in *Frankenstein*. This means that there has to be a certain amount of autobiographical consideration, as her own experience of the Romantics was in many ways a personal one. Consequently the Romantics that are dealt with as individual examples are included only where deemed necessary. As a result, the focus is mostly on Percy Shelley's involvement in *Frankenstein*, as he is arguably the Romantic poet to whom Mary Shelley had the most intimate connection. Fragments of Coleridge's influence are also examined only insofar as they are relevant to her understanding of Romanticism. The examination of Coleridge's *The Rime of the Ancient Mariner*, and its interaction with *Frankenstein*, which is analysed in the second chapter, is used to show how Mary Shelley's criticism of Romanticism does not mean a complete rejection of Romantic principles. Her incorporation of Coleridge effectively highlights the difficulty in negotiating a criticism against an ideology from which she cannot completely distance herself.

In the same vein, there are many different interpretations of *Frankenstein*, from the feminist reading of it being a "phantasmagoria of the nursery" (Moers, 99) or a rewriting of *Paradise Lost*. The validity of these readings is not disputed, and even though each of these interpretations are enlightening and valuable on different levels for different reasons, it would be impossible to synthesise all these elements into the parameters of what this thesis is predominantly concerned with; namely how Mary Shelley is writing a response to idealistic Romanticism which is far from complimentary. This argument will show how *Frankenstein* expresses Mary Shelley's criticism of what Percy Shelley represents ideologically without analysing how it relates to her expectations of domestic bliss, marriage or the implied patriarchal suppression.

Mary Shelley was eighteen when she wrote *Frankenstein* in 1816, and the first edition was published in 1818. Her youth is possibly what lends the novel its energy, and as becomes clear, provides the criticism she articulates with a particular edge and passion. By the time the second edition came out in 1831, much had changed in her life. At eighteen, she had been the lover of a married man. At thirty-four, Mary Shelley was a widow, and with hindsight obviously felt that her criticism had perhaps been too sharp as her rewriting of the text would suggest. Therefore, in

the 1831 edition, there are subtle changes from the earlier edition, most importantly in relation to this thesis, the fact that Victor Frankenstein is subtly altered from being in charge of his own destiny in the 1818 edition, to being a victim of fate in the 1831 edition (Poovey, 340). For this reason, the citations and focus of this thesis has been on the 1818 edition as well as the manuscript edition, which is discussed in the second chapter, as these seem to be closer to Mary Shelley's original vision for *Frankenstein* than her 1831 edition.

The creature that is possibly the most lasting of all of Mary Shelley's characters is often referred to as Frankenstein's monster. As this thesis will argue, the imposition of the term "monster" was not the author's own. As a result, the creature will be referred to as creature or as Victor's creation to maintain the integrity of the novel and to preserve a sympathetic reading of him, which is, as the following arguments will show, vital to understanding precisely how Mary Shelley articulates her criticism of Romanticism.

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## Chapter 1: Questioning Prometheus

Without the aid of the imagination all the pleasures of the senses must sink into grossness.

~ Mary Wollstonecraft, *Letters Written During a Short Residence in Sweden, Norway, and Denmark* (1796), Letter 2.

Mary Shelley's *Frankenstein or the Modern Prometheus* was published in 1818, which places it firmly within the period of the Romantic movement. Mary Shelley's exposure to Romanticism was a deeply personal one as her relationship with Percy Bysshe Shelley, the popular Romantic poet, implies. Even though the writers in this period will not have placed themselves into what is only retrospectively a literary or cultural movement, there was a certain common understanding of what they were doing which is important to consider if one is to appreciate how subtly Mary Shelley dealt with the elements contained within Romanticism. Her use of the ideas contained within Romanticism, and her destabilisation of these ideas, led to the creation of a story that has permeated common consciousness to a point where it verges on being a modern myth. The most important aspects of Romanticism that feature in *Frankenstein* are based on what the Romantics thought of themselves. It is undeniable that Mary Shelley's immediate knowledge or experience of the Romantics and how they saw themselves was informed by her own experience of Percy Shelley and his worldview. As Abercrombie rightly suggests in *Romanticism*, where he tries to define how this ideology was understood by the Romantics, Romanticism was "a certain attitude of mind: an attitude to life" (Abercrombie, 31) rather than a conscious effort to effect literary change. If the Romantic ideology stresses the mind or the working of the mind over realities perceptible by observation, it is logical to conclude that it places its emphasis on what can be simplified as being an inner reality rather than an outer reality. The significance of this is, as Abercrombie elaborates,

a tendency away from actuality. We see the spirit of the mind withdrawing more and more from commerce with the outer world, and endeavouring, or at least desiring, to rely more on the things it finds within itself. (Abercrombie, 49)

The focal point for Romantics lies within the "spirit" of the mind or aspects that spring from that mind; or, perhaps more succinctly put, they were absorbed by the concept of the imagination, and the power inherent in rendering the world around them through that focal point. It is through this focus on the inner self that Romantics found themselves moving away from reality, and therefore perceiving that reality through the locus of their imagination. It is because of this that one can assume that Romanticism is arguably an antithesis to realism. The

glorification of the imagination, which is almost consistently present in those texts now considered crucial or seminal Romantic texts, is what makes the Romantics seem somewhat self-involved in that the inward nature of Romanticism separates it from the real world. It is for this reason that one can see quite clearly how Abercrombie critically jumps to describing Romanticism as taking “its most obvious form in *egoism*. [...] By egoism I mean an inordinate consciousness of self importance” (Abercrombie, 135). Schueller agrees with Abercrombie, but Schueller’s exploration of Romanticism is grounded in his search for a common factor that links what seem to be contradictory elements of this ideology, which is why his expression of this point about egoism is particularly interesting, as it underlines the fullness of this accusation of egoism more dramatically:

Romanticism holds that man’s moral and imaginative powers, not being limited in any sense, can go way beyond what they seemingly can encompass. Thus, the human ego transcends everything and becomes everything. (Schueller, 366)

On first reading, this extract seems to be particularly damning towards Romanticism; however, the value of Schueller’s statement is the fact that it contains the way the Romantics saw this egoism. If one ignores the emotive reaction elicited by the implication that Romanticism is founded on the human ego becoming everything, there is still much to be gained from this extract in relation to understanding the Romantic ethos. The words Schueller uses to describe the process or context in which this arises, such as “not being limited” and his use of transcendence to describe the action performed by the ego, reveal the positive aspects of this self-absorption, this continual gravitation towards the inner self or mind, and it is these positive aspects that the Romantics themselves embraced. These positive aspects result in a world without limitations, where the individual can transcend the mundane and even the human. By envisioning reality in this way, Romanticism does not discard reality, but in the mind of the Romantics, they were in fact “beginning [...] a new reality” (Abercrombie, 117). As Schueller goes on to imply, Romanticism is in its way the drive for a revolution, the yearning for something new, the need to create and transcend outside of the limitations enforced on the individuals by the society around them. This could only be achieved through what they perceived as being the power invested in them by their imaginations, which in turn is why Schueller likens Romanticism, in contrast to Abercrombie’s “attitude of mind”, to something that is perhaps more concrete and yet at the same time more abstract than a specific mind-set:

Thus, Romanticism has some of the qualities of religion: It wants to forsake the world as it is and create a new one: it desires the amelioration of the world’s ills, social, political, moral; the concrete recommendations for change may differ and even contradict one another. (Schueller, 363)

This wanting to create a new world, almost in a religious sense, and their belief that it was within their grasp to do so, also explains why Romanticism produced a form of optimism and idealism. This idealism definitely permeates the literary work of the Romantics, albeit in different concentrations according to the inclination of the individual Romantic. With this emphasis on creation, one can now consider the image of Prometheus in relation to the Romantics, but more specifically in relation to Mary Shelley's use of this image in *Frankenstein*.

By using *The Modern Prometheus* as the subtitle to her novel, Mary Shelley was invoking what could possibly be one of the most revered images or metaphors of Romanticism. The Greek myth of Prometheus exists in two versions, both of which are of interest in regard to *Frankenstein*, not only because her plot draws on aspects of both, but also because the dual nature of this myth was already known by the Romantics, although they definitely favoured Aeschylus's version of events. The Promethean myth, according to Aeschylus, styles Prometheus as a Titan hero of mankind, who stealing fire from the gods against their will, gives fire instead to humanity to give them the means to further their existence. With the gift of fire, Prometheus also has a claim to teaching humanity civilisation, and further than that giving them writing, agriculture, medicine and science. Therefore the term "Promethean" has acquired cultural resonances that stem from this particular myth: namely that it is a way of "referring to defiance of superior powers" when these powers are seen as "opposed to human initiatives" (Podlecki, 1). It is the rebelliousness of the Titan which earned Prometheus a certain heroic status in the minds of the Romantics. This is clearly illustrated by Podlecki in his introduction to *Aeschylus: Prometheus Bound* where he traces the evolution of the myth and the changing receptions of it through history:

Around 1800 Prometheus begins to appear on both sides of the English Channel as a rebel figure, the individual ready to flout convention and challenge entrenched privilege. [...] Prometheus became for the English Romantics an irresistible figure of rebellion, a prototype of the creative artist, isolated, misunderstood, reviled. (Podlecki, 52)

The image of Prometheus incorporates a sense of not only needing change but also actively pursuing it no matter what the cost, to accrue a higher level of independence or knowledge. The irresistibility of Prometheus is that as a mythical figure, he became a sign or a token for the spirit of that age, as Byron wrote in his poem entitled *Prometheus*: "Thou art a symbol and a sign [...] Like thee, Man is in part divine."<sup>2</sup> This again highlights how the creative artist, of which Byron is but one example, saw themselves as being "in part divine" or transcending the normal human

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<sup>2</sup> Byron, *Prometheus* (1816), (III, 45-8)

level through their particular way of focusing egoistically on inner realities rather than actual reality. In fact, the figure of Prometheus was merely lending itself seamlessly to expressing a pervasive arrogance that is articulated in the belief that divinity is an attainable goal for the creative artist. Beer in his explanatory chapter on *Frankenstein* incorporates this image of Prometheus as a rebel yet still divine, more fully in relation to writers of this period by arguing that:

Romantic writers in general, with their concern for human liberty, developed a natural fellow feeling for the great protagonist of humanity. Instead of a Son of God made to suffer by his contemporaries, as promulgated in Christianity, they were drawn to the idea of a man with sufferings directly attributed to his Creator. [...] Why should not they be allowed simply to protest against their plight, retaining in the circumstances a dignity and nobility? Prometheus on his rock became their emblem for such feelings. (Beer, 229)

In a sense, Prometheus was used to articulate the Romantics' notions not only about themselves, but also about what they deemed as their right or their responsibility to do in relation to all spheres of life.

This attitude towards their own society is reflected in their reverence for Prometheus as a rebel, but also relates to the other aspect of the Promethean myth. This image of the isolated creative artist is not only associated with the Titan's theft of fire, but also with Ovid's version of the myth, where Prometheus features more prominently as the creator of man, creating man from clay. As one of Mary Shelley's prominent biographers (Seymour) points out, this role of creator seemed to be yet another aspect of Prometheus not only immediately linked to the spirit of the age, but associated yet again with the literary Romantics in his power of creating a new reality. More important, however, is the fact that it also made the mythical image usable for science:

Prometheus, so the story went, was a god, a Titan who took clay from the plain of Boeotia and from it, moulded man. The secret of creation seemed only a leap away from the grasp of chemists and physicists at the end of the eighteenth century. (Seymour, 4)

This secret of creating a new life is the influential driving force behind Mary Shelley's protagonist, Victor Frankenstein, who as the title suggests, is her modern interpretation of Prometheus. Victor Frankenstein is not only Promethean in his rebellious usurpation of the role of creator, but also sees himself as rightfully transcending boundaries to attain that Promethean ideal.



Therefore, in Mary Shelley's use of the image of Prometheus she draws on both aspects of the myth through her principal character, which as the title suggests is Victor Frankenstein. This use links her novel firmly to the preoccupation of her peers, and consequently Romanticism as a whole. However, the difference between this work and that of her contemporaries is that *Frankenstein* is a cautionary tale, a response to the continual glorification of this isolated artist and creator. Consequently her every use of the Romantic notions or ideas that she herself was so familiar with highlight repetitively that as a whole, Mary Shelley had an unmistakably different view of Romanticism than her peers. There is an inherent critique in her version of the Promethean myth, a darkness that is already apparent in her choice of epigraph:

Did I request thee, Maker, from my clay / To mould me man? Did I solicit thee / From darkness to promote me? (Milton, *Paradise Lost*, X, 743-45)

The agonising question that prefaces the novel already implies a questioning of that right to creation. This epigraph sets the tone of questioning, of implied criticism before the story has even begun. This method of invoking an aspect of Romanticism, such as the image of Prometheus, only to question that aspect moments later, recurs throughout *Frankenstein*. Mary Shelley subtly uses Romanticism, but only to question it. This in essence is why *Frankenstein* becomes a Romantic novel about Romanticism. What is even more appealing or interesting, is that *Frankenstein*, a novel about new creation and a rebellious creator, is undeniably entrenched in a pessimistic view of the Romantic optimism that was familiar to Mary Shelley. It carries with it a warning about the threat of the unbridled imagination, and perhaps beyond that it is a critical look at the creative power of the author or the artist, the responsibility of the creator, and the downfall of the modern Prometheus.

Victor Frankenstein therefore becomes the figurehead of Romanticism, or more specifically of how Mary Shelley saw Romanticism, which is represented in the novel. Through the character of Victor, Mary Shelley has a means to comment on Romanticism, and her use of the image of Prometheus as discussed previously only underlines this connection. As a man of science, he is not as far removed from Romantic principles as it would appear. Wordsworth, in his *Preface to Lyrical Ballads*, linked his notion of the Poet with what he terms the "Man of Science":

If the labours of Men of Science should ever create any material revolution, direct or indirect in our condition, and in the impressions we habitually receive, the Poet will sleep then *no* more than at present; he will be ready to follow the steps of the Man of Science, not only in those general indirect effects, but he will be at his side, carrying sensation into the midst of objects of the science itself. (Wordsworth, 270)

Here it is quite apparent that the Poet will stand side by side with the Man of Science, almost as if the two are inseparable by virtue of sharing the same passion for knowledge, understanding or transcendence. The only difference is that the Poet will be the carrier of sensation, whereas the Man of Science is involved more with the objects of science itself. Victor explains in his narrative of his experiences, that his interest in science was something that he learnt early, but that it was initially only a small interest, and in a sense the fact that this interest went completely unchecked led to the misery that is his later life story:

I must not omit to record those events which led, by insensible steps to my after tale of misery: for when I would account to myself for the birth of that passion, which afterwards ruled my destiny, I find it arose, like a mountain river, from ignoble and almost forgotten sources; but swelling as it proceeded, it became the torrent which, in its course, has swept away all my hopes and joys. (M. Shelley, 22)

This extract is particularly important on two fronts in regards to Mary Shelley's use of Romanticism, as it is an example of how subtly she navigates between Romanticism on the one hand and destabilising it on the other.

Firstly, and here one must digress slightly, this extract is one of many areas where the narrative focuses on Victor's inner reality, his perception of his inner mind as it were, which as discussed previously is more important within Romantic thought than the reality of the outer world. The self-justification inherent through most of Victor's narrative is continually shown to be flawed by Mary Shelley. This focus on his inner reality is not enough, as his inner dialogue never does alleviate the problematic nature or consequences that exist in the real world outside of his own justifications. In other words, the fact that he tries to explain how he arrived at creating his creature, which he articulates in terms which alternately present him as hero and victim, does not alter the reality of the novel.

Secondly, and more pertinently in relation to this particular quote, it shows how Mary Shelley manipulates Romanticism on the level of images and expectations. One of the lasting perceptions of Romanticism is that it was in a sense what can be termed as naturism, or a return to nature. The Romantic according to Schueller is "embracing all of life" and wants to "encompass the world" (Schueller, 365) which in fact is perhaps why so much of Romantic poetry is a return to nature, or trying to find new truth in communing with nature, a drive towards a oneness with nature. Therefore, natural images abound in Romanticism as vehicles to

access that transcendence beyond the human. This focus on nature also relates to the Romantic notion that the isolated artist, standing outside of society, transcends society by embracing nature or life outside of that society. Consequently, notions of nature are usually transcendent or serene. The mountain stream of the extract then, much like other nature images in Romanticism, becomes a vehicle for expressing a certain aspect of Victor's inner consciousness. In contrast to the expected notion of calmness and serenity however, this particular stream is a "torrent" which has "swept away" all the positive aspects of Victor's life. Mary Shelley bends the expectation of a sublime image into a raging reality, rather than a philosophical divinity. What is interesting about her use of this particular image of the "torrent" is that it highlights how the subversion or questioning of the Romantic elements are often expressed by Mary Shelley in Gothic anxieties or tropes, as is the case in this extract. The beauty of nature is replaced with an awe-inspiring yet threatening or devastating nature, as the "raging" implies. This extract also points towards a pessimistic realism that is favoured over the usual optimism and idealism found in Romanticism, showing how minutely Mary Shelley reworks Romanticism towards her own ends.

Mary Shelley remains sceptical throughout *Frankenstein* of the celebration of Romanticism, and consequently her sceptical gaze includes most certainly, science and technology as well as artistic renderings of Romanticism, specifically in written form. However, there is a definite sense in which this criticism includes, more specifically, Percy Bysshe Shelley, but can also include those of his contemporaries who shared his views, which include writers such as Lord Byron, Coleridge and Wordsworth as well as scientists such as Sir Humphry Davy and philosophers like her father William Godwin. Through Victor, Shelley conflates her thoughts on Prometheus, a figure that was held in much admiration by the Romantics, and a Romantic who happens to be a scientist, who thereby represents the "modern Prometheus" accurately. This conflation of Promethean desires into Victor Frankenstein the scientist is an important aspect of Mary Shelley's critique of Romanticism. It is possible that Percy Shelley, being both a literary Romantic and interested in science from a young age, may have influenced Mary into realising that the pitfalls of Romanticism would be given a stronger rendering within the possibilities of having a protagonist who is a scientist rather than a poet. Mellor explains this choice of protagonist by arguing, "the pitfall of the Romantic ego is given a perfect outlet through the scientist yearning for a god-like power of uninhibited creation" (Mellor 2003, 19).<sup>3</sup> Scientists, as has been

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<sup>3</sup> Critics such as Mellor, Hoobler and Badalamenti have illustrated or implied exactly how closely Victor is modelled on Percy Shelley, but for the sake of brevity, I have here assumed this to already be an established fact, and refer specifically to the very simple tabular comparison of Anthony F. Badalamenti, "Why did Mary Shelley write

mentioned, in every way thought that they were moving towards the possibility of creating new life at the time when Mary Shelley was writing. Through her use of a scientist, Mary Shelley can fully explore the literal implications of yearning for a god-like power. Even though Prometheus represents the ultimate Romantic ideal, not only for scientists but more importantly for writers of the late eighteenth century, Mary Shelley's choice to explore that yearning through a scientist, makes the entire critique resound with realism, rather than philosophising about this god-like aspiration in a poet. In her use of Prometheus then, one could argue that it is precisely this pursuit of divinity within Romanticism which Mary Shelley wishes to interrogate.

The knowledge that a human mind can transcend its limitations and attain divinity by creating a new reality through the imagination is expressed in many writings of that time, but in none perhaps as directly as in Samuel Taylor Coleridge's writings. Coleridge's depiction of the artistic, creative act illustrates that the poet only has to harness his creative power to repeat the "eternal act of creation in the infinite I AM" (Coleridge, 202). According to Coleridge and Shelley, in his creativity the poet or literary artist appropriates godlike powers for himself and consequently the poet becomes all-powerful. Mary Shelley seems to have realised that there is a threat or danger inherent in this presumptuous desire for complete and utter artistic power, which is reflected from the outset in her protagonist's longing. Victor Frankenstein longs to create a new "species", but places his emphasis more on the fact that he wants to be blessed as its creator; in other words, the emphasis is on his own overreaching rather than that which he creates, and this is what compels him to continue:

No one can conceive the variety of feeling which bore me onwards like a hurricane, in the first enthusiasm of success. Life and death appeared to me ideal bounds, which I should first break through, and pour a torrent of light into our dark world. A new species would bless me as its creator and source; many happy and excellent natures would owe their being to me. (M. Shelley, 36)

His excitement is propelled by a multitude of motivations, all of which are essentially forbidden to a mere mortal. Victor plans to be the first to break through the ultimate "bounds" between life and death, thereby bestowing on himself the power to create life and thereby defy death, or in Romantic terms, he wants to create a new reality, where he becomes a god. This is also seen only in the positive, as it would in optimistic Romanticism, as he plans to "pour a torrent of light into our dark world" making Victor, in his own mind at least, the ultimate light bringer, or the ultimate personification of Prometheus. These wonders that he foresees are based purely on his

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Frankenstein?", (2006: 426) but the idea of how Victor represents Percy will be developed in detail within the second chapter.

own deification, on his own inner justifications or needs, and the creature he wants to bestow life upon is seen only in relation to the blessing it would obviously bestow on Victor as “its creator and source”. In this short extract Mary Shelley highlights exactly how self absorbed her modern Prometheus has become, focusing only on the glory of becoming the great “I AM”.

This delusion of transcending other mortal beings collapses almost immediately after Victor has created the life that he so longs to be worshipped by. Victor seems to attain Promethean status, but it is once he attains his goal that everything takes a turn for the worse. When confronted with the fruits of his creative frenzy Victor exclaims:

How can I describe my emotions at this catastrophe, or how delineate the wretch whom with such infinite pains and care I had endeavoured to form? His limbs were in proportion, and I had selected his features as beautiful. Beautiful! – Great God! [...] I had desired it with an ardour that far exceeded moderation: but now that I had finished, the beauty of the dream vanished and breathless horror and disgust filled my heart. (M. Shelley, 39)

All his best intentions and “infinite pains and care” result in something that he is not willing to face or acknowledge. Arguably, Victor did not think of his creation but only how it would be a vehicle for his magnanimous creative energy. His idealistic appreciation of his own power vanishes when he is faced with the reality of what he is responsible for. This ties in yet again with the inner reality being incompatible with the outer reality. In Romanticism, the inner reality of the creative mind is lauded as more important than the actual reality in the world, but here Mary Shelley invokes a sort of reality check. Instead of being transported by his divine act of creation, Victor is disgusted by the new life that he has created out of death, by the actual reality of the creature.

However, at this point it is not Victor’s overreaching alone that is criticised, but also his unthinking appropriation of creation or even procreation. Here it seems that because Victor took the act of creation fully on himself, creating the creature without a womb or any female involvement in its birth, that ultimately the creation is counterproductive. Feminist theorists have taken up the fact that Victor creates or in fact procreates without female input on multiple fronts. The fact that he usurps the feminine privilege of motherhood and as a result fails to mother his “child” enhances what Moers calls the “motif of revulsion against newborn life” (Moers, 93). Victor’s denial of the importance of women both in his initial act of creation and in

his refusal to create a female companion for his creature emphasises what Mellor terms a “homosocial theme” or a “bleak parody of Romantic love” (Mellor 2003, 13). Homans interprets this rejection of the feminine by Victor as Mary Shelley portraying the patriarchal Romantic desire to “do away, not only with the mother, but with all females” so that the Romantic’s world can only “reflect a comforting illusion of the male self’s independent wholeness” (Homans, 107). These readings, which are merely a sample of the huge body of feminist criticism that looks at this particular issue, are both valid and compelling; yet to deal with them fully lies outside of the scope of this particular discussion. What is relevant to the present argument is that the creature is actively constructed or made. The artificiality of the act is what makes it unnatural or more precisely the “product of the unnatural coupling of nature and the imagination” (Poovey, 337). The over glorification of Victor’s imagination, which stems from his unchecked ego, results in him not giving any real thought to the necessity of natural procreation. Consequently, it is in this unnatural coupling of ignorance and arrogance that Victor’s downfall is initiated.

Victor’s ultimately disastrous desire to transcend his human limits links him once again to Prometheus as the rebellious over-reacher, and it becomes evident that this disregard for boundaries is deeply entrenched within Romanticism. Victor by breaking one set of boundaries, those between life and death, inevitably breaks another, which is contained in his trying to be both mother and father to his creature. For Schueller, this breaking of boundaries is the one thing that is at the centre of Romanticism as “Romanticism is described as deriving from the urge of the human psyche to go beyond the human confines in which it finds itself” (Schueller, 360). This drive to transcend, to revolutionise, to create anew is explained fully when reduced to its common motivation, namely to go beyond the human confines created by society, religion, and anything else that limits rather than expands the human psyche. As the modern Prometheus, Victor is driven completely by his urge to break boundaries, but sees it only as a duty to bring light, to ultimately become god-like and revered by his creation, and to discover truth or create a new reality, which is based solely on his ego or his imagination. Victor Frankenstein therefore represents the ultimate fall of such presumption, as he has created for the sole purpose of unleashing his own creative power; he has broken boundaries without any sense of possible negative consequences. It is at this point in the narrative that Mary Shelley’s criticism of Victor is perhaps for the first time truly apparent. This unchecked drive to break boundaries or flout convention is something Mary Shelley was familiar with by virtue of living with Percy Shelley. As will be developed fully in the next chapter, Percy Shelley was in many ways the embodiment of

Romantic ideology. The elopement of Mary and Percy in the summer of 1814 is just one of the instances where Percy, a married man, breaks away from what he saw as a limitation on his psyche, in this particular instance his own marriage. The difference between Mary and Percy is that he saw their elopement and consequent relationship mostly as beneficial to his own psyche, as he wrote to a friend:

How wonderfully I am changed! [...] Not a disembodied spirit can have undergone a stranger revolution! I never knew until now that contentment was any thing but a word denoting an unmeaning abstraction. I never before felt the integrity of my nature, its various dependencies, and learned to consider myself as a whole accurately united rather than an assemblage of discordant and inconsistent portions. (in Hoobler, 85)

In contrast, Mary had to deal with less ideal ramifications of what was considered scandalous behaviour. Estranged from her father, growing increasingly irritated with her stepsister who Percy had inexplicably decided to take along, Percy's debilitating debt and her own illness due to a pregnancy are all reasons why during "their first year and a half together, Mary would learn that the dreams of her husband could bring nightmares to her" (Hoobler, 76). The combination of all these factors explain why Seymour states that "Mary was feeling painfully isolated; she had not expected to be so punished" (Seymour, 116) and that Percy's erratic behaviour soon led to the realisation that "she had chosen a man who, like her father, put beliefs before relationships" (Seymour, 119). Here is the first kernel of what would become her first novel. The consequences of following an ideal blindly, as Percy Shelley had done, led to horrifying consequences not only for their relationship but also for herself. Percy Shelley at no point seems to have been aware of the isolation she felt, as he only exasperated it by writing to his wife Harriet to come and join the eloped party (Hoobler, 80). This self-absorption is problematic for Mary Shelley in that the Romantic idealist does not even realise or consider the emotional damage that is done to those around them by breaking through these limitations or boundaries. Drawing on her own experience, Mary Shelley shows in *Frankenstein* how the unchecked drive of Romantic thought leads to horrific consequences. Victor did not foresee or wonder about the horrific consequences that would follow from his breaking through human limitations, for, in true Romantic fashion, his imagined greatness was enough justification for his act.

This reliance of Romantic dreaming of the unrestrained possibilities thrown open by the imagination is another point at which Mary Shelley undermines Romantic thought. According to Poovey, by showing the dangers of an unrestrained imagination Mary Shelley emphasises the fact that "the imagination is properly a vehicle for escaping the self, not a medium of power or even

self-expression. (Poovey, 344). What Poovey is drawing attention to here, is that Mary Shelley is in fact using the failure of Victor's imaginings to highlight yet another fault contained neatly within Romanticism. Schueller more pertinently expresses the same point, highlighting the contradiction inherent in Romanticism, especially in relation to the imagination, as he argues that the difficulty in their concept of the imagination lies in the fact that "the only agency which the human mind has for transcending itself, is itself" (Schueller, 361). He is therefore in agreement with Poovey's notion that the imagination originating within the human mind is limited to barely escaping the self and can therefore not at any point become a "medium of power", or in other words, can not create change in the actual world, nor gain the transcendence the Romantics, or Victor so long for. Victor tries to use his imaginative and consequently his creative powers that originate from those imaginings to create real and positive changes within the world, if not creating a new reality or a new truth. Everything that Victor is hoping to gain from this ends in utter and spectacular failure. His imagination leads him astray as nothing he imagined he would achieve by becoming a creator happens in the way he had wished for, nor does he at any point transcend the reality of his existence, nor is divinity anywhere near his grasp.

Victor Frankenstein's transgression does not merely relate to his arrogant belief in the transformative quality of his own imaginings or creative power. Another point of transgression for Victor, and a critique from Mary Shelley, is that by creating his creature Victor is actively placing himself outside of the rules of nature and society. A denial of death, which in itself is a transgression against nature, bound with a transgression against natural procreation, leads almost directly to Frankenstein's demise, as Poovey argues:

The course of Frankenstein's decline suggests, in fact, that in the absence of social regulation the formation of the ego is primarily influenced by the imagination's longing to deny the fundamental human limitations – in particular the body's determinate bondage to nature and to death. (Poovey, 334)

What Poovey is suggesting here is that the denial of foundational restrictions that are pursued by Victor, in particular the inevitable boundary of death that is part of the natural limitation of life, is judged by Mary Shelley to be something that should not be trespassed against. The slow yet systematic demise of Victor's health is a consequence of an ego unchecked by social regulations. His self-imposed isolation is what allows Victor to give free reign to his inner drive to transcend human boundaries and his deteriorating health and his almost mad pursuit of a divine transcendence is counteracted directly by Mary Shelley's censure, her warning implicitly embedded in the fact that his body is deteriorating as the novel continues. The reader therefore



is forewarned of his demise before it is completed. While Victor is consumed by his need to create life out of death, to become a god, Shelley continually draws attention to Victor's health, and his relationships with those around him. The dismal state of both act as a warning as he is transported into an ever increasing frenzy by what he imagines, but more importantly when he actively pursues these imaginings with the intent of transposing those imaginings into the reality of the world:

Winter, spring and summer, passed away during my labours; but I did not watch the blossom or the expanding leaves – sights which before always yielded me supreme delight, so deeply was I engrossed in my occupation. [...] Every night I was oppressed by a slow fever, and I became nervous to a most painful degree; a disease that I regretted the more because I had hitherto enjoyed most excellent health, and had always boasted of the firmness of my nerves. But I believed that exercise and amusement would soon drive away such symptoms and I promised myself both of these, when my creation would be complete. (M. Shelley, 38)

Before this point, Victor “had hitherto enjoyed most excellent health”, and it is only in the actualisation, in other words, when he is taking steps towards achieving what he has imagined when he starts creating his creature that his health deteriorates. Therefore, it is not Victor's imagination in itself that Shelley seems to condemn, but only his unchecked egotism of believing that a positive change can be effected by his creative act. Consequently, it is with this act of creation that Victor Frankenstein is condemned for the presumption that this change that he has imagined will be for the enhancement of society. Allowing oneself to actualise the literal implications of an unchecked imagination, in other words indulging the ultimate ideal of Romanticism, is to invite tragedy, and ultimately for Shelley this can only end in death. As Mellor points out:

The modern Prometheus steals the “spark of being” from mother Nature. As Victor works, lightning flashes around him, storms rage on land and sea, rain falls. For Mary Shelley the penalty for penetrating into the recesses of nature, to pursue her to her hiding places is death, in this case the curiously *natural* death of Victor Frankenstein at the young age of 25. (Mellor 2003, 19)

Therefore, this transgression against the natural order, his attempt at becoming a creator of his own species, ends in the significant eliminating of his own spark of being, not by the hands of his creation, but almost inevitably through a natural death. The fact that this deteriorating condition ends his existence only reinforces Victor's obliviousness to the warning signs contained within his ailment. Arguably, therefore it is his own awareness of his waning state that makes him less of a victim and more of a fool for completing his creative act at all. If he had died in any other way, there would be some doubt as to what Mary Shelley is trying to illustrate. The almost anticlimactic nature of his slow, drawn out suffering is evidence that the only one to

blame for his demise is Victor himself. In contrast to Prometheus, Victor does not suffer by being let to live eternally chained to a rock that continually reminds him of his transgression, for that would have meant a partial success at attaining Promethean qualities. By killing him slowly yet naturally after an extended illness, Mary Shelley underlines how profoundly human and mundane Victor Frankenstein remains even after his creative act, undermining his desire to transcend his human body right up to the point where he draws his last breath.

However, it is not merely the fact that Victor Frankenstein transgresses against nature by trying to transcend his human state that Mary Shelley is criticising. Linked to this preoccupation of seeking divinity is yet another tenet of Romanticism that Mary Shelley interrogates. By trying to transcend the human, by pursuing self-deification, Romanticism celebrates solitude, for it is only in the complete isolation from the human and consequently from human society with all its implied regulations and boundaries, that the Romantic can transcend into something that is beyond the human. As Schueller illustrates:

[Romanticism] is a phenomenon directed by the urge to break apart the bonds of existence here and now, to extend existence for the individual beyond the confines of the twenty or twenty-five persons each of us is fated to pass our lives with. The Romanticist wants his life to expand beyond these confines. (Schueller, 363)

Again, Schueller draws attention to the fact that the Romantic wants to break boundaries to pass into something outside of the norm. The confines that are represented by the people surrounding the Romantic, or in other words, society as a whole, is just another constraint that needs to be broken away from. Romanticism therefore regards complete isolation as desirable as community and family only restrain the Romantic. Consequently, in Romantic ideology it is only through solitude unhindered by others that something new can be created.

The solitude that is portrayed throughout *Frankenstein* is a continual theme in the narrative not only of Victor Frankenstein, but also in the narrative of Walton, as well as that of the creature. In criticising the Romantic ideal of the imagination leading to a romanticised transcendence, Shelley highlights the antisocial aspects of isolation rather than the triumphant break from human limitation. Victor veers into madness, only because he completely rejects all his needs for community or family. His self-asserted isolation at one point even includes the forces of nature, showing in the same instance how far he has digressed from sanity, but also how complete his isolation has become:

Oh stars and clouds, and winds, ye are all about to mock me: if ye really pity me, crush sensation and memory; let me become as nought; but if not depart, depart and leave me in darkness. (M. Shelley, 122)

Victor has become so far removed from society that he speaks to the nature around him as if even that company is too much for him. This leaves him ultimately stranded in a state of utter loneliness; the solitude he has actively chosen for himself no longer has the feverish anguish of needing to finish his work in peace, but now turns towards a need for isolated darkness, which highlights his crippled sense of society rather than elevating him to new heights. Strangely enough, it is this recurring theme of lonely solitude that is often critically read as a positive element that serves to reinforce Romanticism, as Ferguson aptly demonstrates in *Solitude and the Sublime*:

In a novel that celebrates both solitude and friendship, that is, solitude itself is both friendship and recommendation for friendship. Even Victor's apparently solitary appreciation of nature (as a respite from care, as a departure from society) tends to look less like the kind of pathetic fallacy of Romanticism [...] (Ferguson 1992)

Ferguson's take on *Frankenstein* sees Victor's increasing isolation as a reinforcement of the Romantic ethos; as he states, it is an "amiable isolation". What is particularly erroneous about Ferguson's reading of *Frankenstein* is that, like Victor Frankenstein, he only sees the celebration of isolation and consequently the bringing of enlightenment through that isolation that underlines Romantic ideology. This contradicts the fact that Mary Shelley is criticising precisely the presumption that solitude should be celebrated. One can only see Victor being a figure of celebration if one completely ignores the many subtle ways in which Mary Shelley undermines her modern Prometheus. One example of this is in the ironic way in which Victor Frankenstein, who aspires to be the bringer of light in the Promethean sense, is followed or consumed by darkness throughout the novel. What Ferguson terms as a joyous "departure from society" is consistently undercut with darker images, which seem to indicate a very clear emphasis on the stark loneliness or darkness in Victor's solitude in contrast to the anticipated light or divinity that he wants to acquire. Even when he is in the first throes of his scientific research, there is a macabre juxtaposition of Victor's enjoyment of his solitude and the dark reality of his scientific search:

Darkness had no effect upon my fancy; and a church-yard was to me merely the receptacle of bodies deprived of life, which from being the seat of beauty and strength, had become food for the worm. Now I was led to examine the cause and progress of this decay, and forced to spend days and nights in vaults and charnel houses. My attention was fixed upon every object the most insupportable to the delicacy of the human feelings. I saw how the fine form of man was degraded and wasted; I beheld the

corruption of death succeed to the blooming cheek of life; I saw how the worm inherited the wonders of the eye and brain. (M. Shelley, 34)

This extract is only one example of how Mary Shelley subtly maps out the tensions inherent in Victor's research, all the while highlighting his preoccupation with his studies. Even though the extract draws attention to the excitement that Victor has in acquiring so much knowledge, there is no denying that it is shadowed by the repeated images of darkness. This darkness is present in all the places he frequents, the cemetery and "charnel houses", the "bodies deprived of life" which are "degraded and wasted" and his fascination with the "corruption of death". This immersion in darkness degrades his solitude into something more sinister as he is shadowed by not only the death that surrounds him, but the inevitable foreshadowing of his own death.

Victor's isolation is not only blackened by a consistent link to storms and darkness but is also in many ways linked to his untimely yet inevitable death. His survival, dragged out as it is, needs to be remarked upon; his lingering does nothing to alleviate the tragedy he has put into motion, and his eventual death underlines the severity of Shelley's critique of his actions and his character. His delayed death is quite simply used as a narrative device. Alwes, who draws links between *Frankenstein* and what could be termed seminal Romantic texts in terms of Victor's alienation, argues as follows:

Victor is left alive once again, (like the Ancient Mariner, he survives all deaths in the novel, until his own, in order to tell the tale), alienated and impotent outside the growing circle of the accumulating dead. (Alwes, 113)

This link to the Ancient Mariner from Coleridge's poem is an important one, as it draws attention to the fact that it hauntingly echoes into Shelley's own understanding of solitary existence as potentially alienating. Walton is another example with which the Ancient Mariner image is reinforced in *Frankenstein*. Walton being a sailor is an obvious link, but in addition he states that he is "going to unexplored regions to 'the land of mist and snow'; but I shall kill no albatross therefore do not be alarmed for my safety" (M. Shelley, 10). The killing of the albatross is an obvious reference to Coleridge and as Alwes explains:

[It] reinforces Walton's own solitariness, as the Ancient Mariner is product and prototype of Romantic alienation and, further, of loss of salvation, a theme pertinent to *Frankenstein*. Whether enforced or invited, solitude is the only reality in the novel, and with the concurrent loss of each character who tries and fails to find community within family, solitude becomes akin to solipsism. (Alwes, 112)

Walton seems to identify throughout with Victor, even after conversing with the creature himself, as this extreme preoccupation with or indulgence of one's feelings or desires is something that they have in common. For this reason, their solitude becomes "the only reality in the novel". The continual reference to darkness and solitude is also enacted by Walton, who in his first letter to his sister writes:

I try in vain to be persuaded that the pole is the seat of frost and desolation; it ever presents itself to my imagination as the region of beauty and delight [...] Its productions and features may be without example, as the phenomena of the heavenly bodies undoubtedly are in those undiscovered solitudes. (M. Shelley, 5)

Walton then, like Victor, is drawn to the exploration past what are seen to be boundaries of knowledge; in his case, however, the boundary is geographic rather than that between life and death. For Walton, as with Victor, the allure of breaking through these boundaries is motivated by a desire for fame and to be remembered by future generations; he wants to be acclaimed as a bringer of knowledge just as Victor does. In his very first letter to his sister he writes a motivation for his journey, explaining that "you cannot contest the inestimable benefit which I shall confer on all mankind to the last generation, by discovering a passage near the pole" (M. Shelley, 6). Victor uses a similar turn of phrase later in the novel as the central motivation for breaking through boundaries perceived to be unbreakable. Yet somehow, Walton manages to outlive both Victor and Victor's creation. At the end of the novel, Walton watches the creature "borne away by the waves, and lost in darkness and distance" (M. Shelley, 191). Mary Shelley therefore ends the novel not with Walton's quest coming to fruition, but rather with this disappearance into the "darkness and distance", which leaves Walton ultimately alone, as Alwes argues:

The "darkness and distance" that now lie between the monster and Walton effectively manifest Walton's newfound lack of desire for community and dismissal of the desire for masculine knowledge, characterised by solitude, that return Walton to the aforementioned culture. Throughout the narrative between the monster and himself, concluding in an admonition of the monster by Walton, it is clear that Walton's sympathies lie with Victor Frankenstein – the solitary creator, usurper and scientist – rather than with his creation, who desires above all community with others and abjures his self-taught knowledge. (Alwes, 116)

Walton's quest or "desire for masculine knowledge" is not fulfilled, and he remains in solitude. It becomes unimportant whether or not he succeeds, as the emphasis falls on the fact that the creation "who desires above all community with others" is left sadly alone. It is not the solitude of Walton which entices a sad reaction to the ending of *Frankenstein*, as Walton is in effect not completely isolated. His letters to his sister are one link to society that he does not sever, and as

tenuous as this link seems, it ultimately results in his survival, the implication being that he will return home and therefore he can redeem himself in a way that Victor refuses to do. The reader's sympathy at the end of the novel is therefore predominantly with the creature's plight as he is the only one of the three narrators who wishes to be included in society but is left with no choice but perpetual solitude in the eternal ice steppes close to the North Pole.

Focusing on the creature then, it becomes important to ask the one question which is repetitively asked throughout *Frankenstein*, through both Victor and his creature, and arguably on which the entire criticism of the Romantic ethos hinges; namely: "Is the creature frequently referred to as "Being" innately good or innately evil?" (Mellor 2003, 20). Continually during the creature's narrative, there is an emphasis by Mary Shelley on Berkeley's philosophy that to be is to be perceived. Any knowledge of the world around can only be obtained through direct perception (Berkeley, 114). This links to the importance in Romantic thought of perception, seeing or visions as being the transcendent truth of reality. This particular emphasis on perception is exposed as erroneous or often leads to a misjudgement of reality, as is shown by the reaction of others to the creature. This is underlined by Mellor's explanation of the social context that the creature finds himself in:

In the eighteenth Century physical appearance was deemed to be inextricably linked to the inner being, in other words physiognomy and character are closely related. Therefore, throughout the novel, the creature's encounters with humanity are consistently overshadowed by the assumption that he is a threat by virtue of his outer appearance alone. (Mellor 2003, 21)

The fact of the creature's monstrous appearance leads everyone he encounters to immediately assume that his character is as repulsive as his appearance. Mary Shelley's description of the creature and the reactions of others, most importantly Victor's reaction to it, link the creature rather strangely to the Romantic notion of the sublime. The sublime is deliberately invoked to inspire awe, deep reverence or lofty emotions in the reader because of its beauty, vastness or grandeur.<sup>4</sup> The aim was to entice the reader to transcend the normal range of emotions, to attain "the higher degree of everything achieved by whatever means" (Schueller, 360), to encapsulate that grandeur where words would usually fail. This enticement, if successful, would allow the reader to access a new truth that exists beyond the limitations and boundaries of the normal everyday existence. In the case of the creature however, his uniqueness, while having all the elements of being sublime, is construed to be evil, threatening or dangerous rather than

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<sup>4</sup> Definition acquired from the *English Oxford Dictionary*.

transcendent. The creature puts a pessimistic spin on the idealistic Romantics view of the sublime, which by virtue of being idealised should always tend towards the transcendent rather than the threatening. Mary Shelley seems to be pointing out that when one is truly confronted with something that passes all understanding, humanity does not wait to be enlightened by it, but rather without question consistently reacts with aversion. This pessimistic view of the idealised notion of the sublime is yet another way in which Mary Shelley questions the validity of Romanticism. It is often misunderstood by the reactions of those around him that the creature therefore is in fact monstrous, as Poovey demonstrates by her statement of the “monster’s” self-consciousness being linked to the acknowledgement of its own repulsiveness:

For the monster, self-consciousness comes with brutal speed, for recognition depends not on an act of transgression but only on literal self-perception. An old man’s terror, a pool of water, a child’s fear are all nature’s mirrors, returning the monster repeatedly to its grotesque self. (Poovey, 337)

One can agree with Poovey that the instances she lists are indeed all instances where the creature can reflect on his appearance or how others perceive him; however, there seems to be more than a repeated emphasis not on his grotesque self, but rather on the swiftness of others to perceive him as evil. The old man’s terror that Poovey refers to occurs only in conjunction with the reaction of his family seeing him talking to the creature. Before that point, as he is in fact a blind man, the old de Lacy father was listening to the creature’s reasoning with sympathy. After the creature exclaims that he is indeed “an unfortunate and deserted creature; I look around, and I have no relation or friend upon earth” (M. Shelley, 108), throwing himself on the mercy of de Lacy’s hospitality, de Lacy responds reassuringly:

Do not despair. To be friendless is indeed to be unfortunate; but the hearts of men, when unprejudiced by any obvious self interest, are full of brotherly love and charity. (M. Shelley, 109)

The emphasis of the old man’s response focuses on the “brotherly love and charity” inherent in all men. The only inhibitor for such expansive understanding is “obvious self interest” which highlights Mary Shelley’s own continual emphasis on the fact that indulged self-interest lies at the very heart of this tragic tale. The wrong perceptions of those around the creature, and above all, the incorrect assumption made by his creator, Victor, who is too self-absorbed to respond to the creature’s pleas sustain the tragedy of *Frankenstein*. Mary Shelley therefore adds another evil to the ever growing list of dangers inherent in the denial of all else in the face of a selfish quest for knowledge, transcendence and divinity, fuelled by a fevered and unencumbered imagination. The societal comment implicit in the continual rejection of the creature is indeed that people are threatened by what they do not know, and therefore undermines this continual drive for new and

extended knowledge. Essentially, it is not the knowledge in itself that is harmful, but the way in which it is pursued ruthlessly and without pause, and how ultimately such knowledge is executed. The creature therefore is the victim in that he did not ask to be created, and even though he tries to be “innately good”, his continual wish being for companionship and community, he is continually driven to violence by the complete rejection of everyone around him. The answer for Mary Shelley then as to whether the creature is innately good or evil is not as simple as just placing him into one or the other category. He was created with a possibility of being good or of being evil, and her criticism lies not so much with the creature’s actions as with those of Victor and society who abandon the creature; their reactions towards him fuel his own actions.

In light of the tragic events that follow from the creation of Victor’s creature, it is important to realise that Mary Shelley’s criticism of Romanticism is crystallised in Victor’s failings. Victor as the modern Prometheus, or in other words Victor as the embodiment of Romanticism, fails spectacularly to attain the glory, lasting fame or divinity that drives his quest for knowledge past natural boundaries. His Promethean yearnings are not realised, nor does he live on after the end of the novel; he is denied even that fleeting immortality. He transgresses against nature and society, and therefore unleashes a darkness rather than enlightenment into the world. His creature does not embody this darkness, but rather it is a direct result of Victor’s complete self-absorption, his complete lack of taking responsibility for his creature; ultimately it is his failure to perceive any reality or truth outside of his own inner truth or mind. He ostracises himself from society as one would expect a Romantic to do; but neither he, nor society, nor generations to come are enriched by this alienation. Rather, he dies, slowly and painfully without grandiose spectacle at the age of twenty-five, having squandered his life on something that fundamentally does not come to fruition. One cannot doubt the fact that Victor is at fault; it is Victor the scientist who is the perpetrator rather than his creature. The creature, in contrast to Victor, is driven only by his need for companionship, which is rejected by society who sees him as a threat due to the form Victor unthinkingly bestowed on him; and finally it is Victor again, who from the outset abandons him, denying his desire for companionship. The creature is therefore consistently wronged, and even though Victor sees him as a monster, when seen from this perspective and considering all that Victor has done, one must reach a completely different conclusion. Mary Shelley’s consistent criticism of Victor as he embodies each aspect of Romanticism as this chapter has discussed, implies rather that the monster is in fact Victor. However, it is my contention that Victor is criticised precisely because he is the incarnation of



idealised Romanticism. Therefore, one could argue that it is ultimately Romanticism that is undermined by Mary Shelley and consequently is at fault, and as a result, it is Romanticism that is altogether the overarching and utmost monster or monstrous element contained within *Frankenstein*.

University of Cape Town

## Chapter 2: Percy Shelley's interaction with *Frankenstein*

I weigh not what ye do, but what ye suffer,  
Being evil. Cruel was the power which called  
You, or aught else so wretched, into light.  
~ Percy Shelley, *Prometheus Unbound*, (I, 480-2), (1819)

As established in the previous chapter, Mary Shelley wrote *Frankenstein* as a cautionary tale, skilfully undermining some of the cornerstones of radical Romanticism. Using Victor as the expression of her own interpretation of the “modern Prometheus” allows Mary Shelley to highlight the threat of an idealised Romantic ideology. Victor aptly shows the danger of solitude as he reverts to narcissistic solipsism and alienation, and his unfettered imagination inevitably leads him to unleash onto the world something for which he is not prepared to take responsibility at any point. Mary Shelley illustrates repeatedly that overreaching oneself leads ultimately to tragedy and, in the specific case of Victor, to a premature death. Due to the nature of his demise, one can argue that Victor's transgressions are, to Mary Shelley, unforgivable. It is precisely this Promethean trait of overreaching that makes Victor the epitome of the Romantic hero, who is driven by his own estimation to enlightening the generations to come. Yet Mary Shelley judges him as having failed ultimately in the reality of the world. Percy Shelley, without stretching the imagination, is also the radical Romantic hero that Victor represents, even if only in his own mind or estimation. Hoobler summarises Percy Shelley's temperament:

Shelley often saw himself as a solitary genius or a wandering poet, but though he would roam, he also liked to be the centre – and leader – of a group. Indeed, his life and career were devoted to bringing others along with him toward his envisioned, more perfect existence (Hoobler, 54).

Within this description of Percy Shelley, it is clear that he saw himself as that hero the Romantics strove to be: a “solitary genius” who was working towards his “envisioned more perfect existence”. In this description, Hoobler touches on two vital aspects of celebrated Romanticism that Percy Shelley exhibits, namely that his isolated roaming state leads unquestionably to genius and that he envisioned a perfectible state of being that he needed actively to work towards. This description of Percy Shelley illustrates that he was a poet immersed in Romanticism, and therefore similar to Victor Frankenstein who is used to exhibit similar Romantic traits. The important aspect of Percy Shelley's adherence to Romantic principles is that he “saw himself” as responsible for others, a Romantic hero as the “bringing others along” implies. Percy Shelley was also consistently interested in alchemy, chemistry and specifically electricity from his early school

days. His “electrical kites” expressed the particular attraction electricity had for Percy Shelley, and apparently, once when he was home from school on vacation he offered to treat his sister’s chilblains by “electrifying” her (Hoobler, 57). This fascination with electricity as a “cure” is repeated by Victor Frankenstein’s use of electricity to “cure” death. However, it is not merely the attraction to electricity that links Victor Frankenstein and Percy Shelley. Mary Shelley’s naming of Victor is significant here because Victor was one of Percy Shelley’s earliest pseudonyms for himself when he started publishing. Fascinatingly, “Percy had used Victor as a nom de plume for some of his youthful poems, and “the Victor” with a capital V, is also frequently used in Milton’s *Paradise Lost* to refer to God” (Hoobler, 155),<sup>5</sup> thereby seemingly conflating Victor’s link to Percy, and to the need to be revered as a creator, or in other words, his Promethean sensibility. One could argue that by using the name Victor, Mary Shelley is connecting not only Percy Shelley and her protagonist, but is also expressing Victor Frankenstein’s desire for godlike status and perhaps is commenting on a particular character trait of Percy Shelley that she could not express directly. However, the drive towards self-deification Mary Shelley expresses through Victor is an important aspect of literary Romanticism. It would not be far fetched to assume that Percy Shelley, like Coleridge, saw the creative aspect of his poetry on par with the godlike creation of Prometheus or any creative deity. His own choice of the name Victor, specifically in context of Milton’s *Paradise Lost* which he was definitely familiar with, implies that Percy Shelley saw himself not only as the isolated genius of Romanticism but also sought to express that part within himself that was divine by virtue of being a creator.

The similarities between Percy and Victor are based on the fervent nature of their involvement in Romantic ideologies as shown above, but are furthermore consistently mapped by Mary Shelley throughout her novel, on a very personal level. Percy Shelley, like Victor, grows up in an aristocratic family; and within that family, they both have a sister named Elizabeth whom they particularly favour. Victor immerses himself in science to unearth and then finally conquer the boundaries between life and death. Percy Shelley was similarly absorbed by scientific pursuits, spending as much as ten years of his life experimenting with electrochemistry and biochemistry (Badalamenti, 426). Percy Shelley’s fascination with tombs or gravesites is also apparent in the fact that the consummation of the love affair between Mary Shelley and Percy Shelley, according to Seymour, is said to have taken place at Mary Wollstonecraft’s gravesite. This would have been

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<sup>5</sup> “the almighty Victor to spend all his rage” (Milton, *Paradise Lost*, II, 144), “Their fight, what stroke shall bruise the Victor’s heel.” (Milton, *Paradise Lost*, XII, 385) These are merely two examples, from the beginning and the end of *Paradise Lost* to indicate the consistent use of Victor as a name of god.

the only place they were without a chaperone and the story both told of their elopement is that they declared their love for each other at this gravesite after which they decided to elope. It is therefore as Seymour implies:

The discreet north-eastern corner of St. Pancras churchyard would have seemed an appropriate setting, as if Mary Wollstonecraft were presiding over their union. Her grave was conveniently shaded by willows. (Seymour, 93)

One can only speculate how Mary Shelley felt about the creation of life in such close proximity to her mother's last resting place, especially in view of the fact that the child conceived there died very soon after being born in 1815. The death of her firstborn is important as it leads to the first schism between Mary and Percy. According to Mellor, Percy Shelley was unsympathetic to Mary's need to grieve, as he barely seemed to notice it, and appeared to be more concerned with his own affairs. Mellor points out that the day after the baby's death:

Percy [was] gone again, despite Mary's depression. We see already a pattern that would recur. Percy Shelley seems to have been singularly unconcerned with the welfare of his female children, and unmoved by their deaths. He clearly did not share Mary's grief. (Mellor 1989, 32)

One could argue that Mary Shelley's realisation or resentment of Percy Shelley's selfishness originated from this point, quite early on in their relationship. That Mary Shelley felt some form of resentment towards Percy Shelley is arguably illustrated by Victor's death. Victor Frankenstein dies at the end of the novel, at twenty-five, the same age Percy was when Mary finished her novel. This parallel allows an empathetic insight into Mary's feeling of abandonment inflicted by Percy, which is echoed in *Frankenstein* by Victor's rejection of his creation. The lack of paternal affection that Victor shows towards his creation is perhaps also an imitation of the lack of feeling Percy showed towards the death of his first child.

The correlation between Percy Bysshe Shelley and the fictional character of Victor Frankenstein takes an interesting turn when we consider Percy Shelley's involvement in editing Mary Shelley's original manuscript. With all these seemingly obvious connections between her lover and her misguided, narcissistic protagonist it is astounding that Mary Shelley, as she was writing *Frankenstein*, would give her finished pages to Percy to edit them at all. In addition, it is even more surprising that Percy Shelley did not see the parallels that existed between Victor Frankenstein and himself. He consequently also seems to have also missed the implied critique to his own character to the point where he identified quite strongly and undeniably with Victor as his editing proves. The original manuscript of *Frankenstein* survives in the Abinger Collection

in the Bodleian Library, and even though most critics assume that Percy's corrections were for the better, Mellor argues that not all of his corrections were unbiased and that at some points he obviously "misunderstood his wife's intentions and distorted her ideas" (Mellor 1989, 59). It is these particular changes, which are of most interest when looking specifically at the identification of Percy with the novel's protagonist. As Mellor points out:

Percy Shelley on several occasions actually distorted the meaning of the text. He was not always sensitive to the complexity of the character created by the author. He tended, for instance to see the creature as more monstrous and less human than did Mary. (Mellor 1989, 62)

This in itself, his tendency to see the creature as "more monstrous and less human" than Mary Shelley intended, gives the first indication that Percy Shelley seemed to be seeing the monster in the same terms as Victor sees him, rather than seeing him dispassionately or even sympathetically. The fact that Mary Shelley saw the creature only as potentially monstrous rather than an actual monster is still embedded within the text. Mellor argues that Percy left the narrative of the creature, when it explains itself to Victor, intact, and it is here, unsullied by Percy's editorial prejudice that Mary Shelley's voice is consequently still strongest. When first confronted with his creation, Victor is aggressive and sees him as a threat. The creature cries out emphatically, in Mary Shelley's unedited words:

Remember, thou hast made me more powerful than thyself; my height is superior to thine; my joints more supple. But I will not be tempted to set myself in opposition to thee. I am thy creature, and I will be even mild and docile to my natural lord and king, if thou wilt also perform thy part, the which thou owest me. Oh, Fankenstein, be not equitable to every other, and trample on me alone, to whom they justice and even thy clemency and affection, is most due. Remember, that I am thy creature: I ought to be thy Adam; but I am rather the fallen angel, whom thou drivest from joy for no misdeed. Every where I see bliss, from which I alone am irrevocably excluded. I was benevolent and good; misery made me a fiend. Make me happy, and I shall again be virtuous. (M. Shelley, 77-8)

This anguished appeal is very important in regards to what Mary Shelley was trying to set up between the character of Victor and his creation. Here the creature speaks eloquently and calmly, surprisingly so if one takes into account Victor's reaction to him. The creature gives Victor the acknowledgement that he craved when he embarked on this creative enterprise, namely to be "blessed" as a creator, or god-like. In the first sentence of this extract, the creature seems to be praising Victor's artistry by pointing out that "my height is superior to thine; my joints more supple". This has a dual effect: it commends Victor for having made a creature that is above mortal strength, but one cannot deny that inherently there is a threat in this, reminding Victor that his creation is far stronger than he is by his own design. This effectively brings together the

double-edged commentary on Victor's character, as he himself designed the enormity of the physical threat that is posed by his creation, but it also reiterates the unthinking process which Victor employed to see his imaginings realised. He has, however, succeeded in one way, in that the creature lauds him as his "natural lord and king". Following that, the creature points out that he does not want to be seen in opposition to Victor because he is "thy creature, and I will be even mild and docile to my natural lord and king". Here is the essentially human quality of the creature brought to light. He has no need to be seen as the enemy, he does not even question Frankenstein's place as his "king" and creator; all he asks, is that Victor remembers that responsibility, which he movingly cites as that part "the which thou owest me". Here Victor's Romantic ambition is realised, his creation does see him as his lord, yet Mary Shelley shows how the egocentric nature of the Romantic refuses to accommodate that which is owed in consequence. This reasonable entreaty goes against everything that the reader has been led to expect from the creature since Victor's only reaction to him is one of horror and disgust. This appeal to Victor's humanity is not the howling of a monstrous creature, but rather is the pleading of a creation who has been wronged by his god.

This notion is further underlined by the fact that at the end of this speech act, there is an embedded religious subtext as he says: "Remember, that I am thy creature: I ought to be thy Adam; but I am rather the fallen angel, whom thou drivest from joy for no misdeed." Here Victor's ultimate failure is condensed into a single, powerfully emotional line. He has succeeded in making the creature beyond mortal strength and physically intimidating, but has completely ignored his creation's need for emotional and paternal support. There is no doubt that Mary Shelley intended the creature to be read with empathy at this point. This haunting plea is important in establishing sympathy for the creature, which up to this point in the narrative has been perceived by the reader only through Victor Frankenstein's lack of understanding. In the creature's narrative, Victor's guilt becomes most apparent. Percy Shelley, in his editing, left large tracts of the creature's narrative untouched, focusing instead on "undercut[ting] her otherwise consistent portrayal of Frankenstein as an egotist who perceives only his own his own feelings and dangers" (Mellor 1989, 63). As Mellor highlights the changes Percy Shelley made to the original manuscript, one gets the impression that he assumed that the creature's narrative was not important or threatening in itself, consequently leaving it untouched. This is interesting because this maintains that which undermines Victor's Promethean status the most. It is also precisely the point at which the narrative evokes an emotional response in the reader. It is only

through the sympathy induced by Victor's creation that Victor's monstrosity becomes even more evident. Therefore, Percy Shelley leaves the creature and therefore Mary Shelley to express the abandonment and feeling of rejection without his interference, which is remarkable exactly because everywhere else in the text he went to great pains to increase the binary opposition of Victor and his creation, making Victor seem less to blame and the creature by contrast as consistently monstrous. It seems that there is a certain sense in which Percy, as a Romantic, underestimated the powerful expression that survives in the creature's narrative. In his mind Victor was clearly the only figure who commanded sympathy from the reader, as he himself identified strongly with Victor as a misunderstood Romantic genius. The irony of his interference is that it potentially enhances the sympathy that is felt for the creature, as Victor fails to comprehend the human need of the creature for companionship.

Even when Victor breaks his promise to finally fulfil his duty towards his creation, aborts his second act of creation before it is completed and consequently fails to create a companion for the lonely creature, Percy Shelley's editing places emphasis on the monstrous nature of the creature rather than the selfish scientist:

When Frankenstein destroyed the female creature, and Mary had the creature withdraw "with a howl of devilish despair," Percy added "and revenge," thus blunting our sympathy for the forever forsaken creature and destroying the author's more perceptive understanding of the monster. When Mary wished to stress the creature's identification with Frankenstein by assigning the word "wretch" to them both within four lines, Percy changed the second "wretch" to "devil", thus implying that the creature is in fact more reprehensible than Frankenstein. And it was Percy Shelley who introduces the oft-quoted description of the monster as "an abortion", a term he again applied to the creature in his unpublished review of *Frankenstein*. (Mellor 1989, 62)

This change is important because by actively interfering with the text in this manner, Percy Shelley is in fact slanting the sympathy of the reader towards a more approving reading of Victor Frankenstein and a condemnation of his creation. This alteration is thus a direct contrast to the more subtle interchange between them that Mary Shelley had intended. Adding revenge, which is not in Mary Shelley's initial rendering of the creature's reaction, Percy Shelley yet again enhances the threat of the creature; "blunting", as Mellor rightly points out, the empathy inherent in the breaking of a promise which was meant to rectify his initial abandonment of his creation. Victor himself, upon hearing the initial outcry for empathy from his creation, states that:

For the first time, also, I felt what the duties of a creator towards his creature were, and that I ought to render him happy before I complained of his wickedness. These motives urged me to comply with his demand. (M. Shelley, 79)

Here as Victor acknowledges he “for the first time” realises his “duties” as a creator, and that consequently the creation’s happiness is in direct relation to his rendering; in other words, his responsibility as creator is not just to create but then also to take an interest in the needs of his creature. Therefore, because of this acknowledgment or realisation, a possibility for a reprieve between Victor and his creation is brought into consideration as a potential outcome of the story. This possibility exists as a result of Victor realising that his creature has a desire and a need that lies outside of his own.

This line of argument is vital to understanding precisely how Percy Shelley misunderstood the text that he was editing, for in those parts that are left unedited by Percy Shelley, there is a consistent implication of possible redemption for Victor. Percy Shelley did not alter the possibility that is retained in the text by virtue of Victor’s acknowledgement of his responsibility and it therefore remains an integral part of the story. For Victor, this acknowledgement is as close as he gets to stepping away from the solipsistic aspect of his nature that Mary Shelley is commenting on, and yet his unwillingness to create the female creature as a companion undermines his “motives”, as does his continual revulsion for what he has created:

His words had a strange effect upon me. I compassionated him, and sometimes felt a wish to console him; but when I looked upon him, when I saw the filthy mass that moved and talked, my heart sickened, and my feelings were altered to those of horror and hatred. I tried to stifle these sensations; I thought, that as I could not sympathise with him, I had no right to withhold from him the small portion of happiness which was yet in my power to bestow. (M. Shelley, 121)

In these words, untouched by Percy Shelley, Mary Shelley paints Victor as torn between the visual horrors of what he has created, and compassion for what his creation is saying and asking for. Victor acknowledges that “I had no right to withhold from him the small portion of happiness which was yet in my power to bestow.” This sentence stands in direct contrast to Percy Shelley’s notion that Victor was a victim of evil. As a faultless victim, there would be no point in the narrative at which Victor would be moved by his creation’s plea as this extract implies he in fact is. The phrase “I had no right” is more in line with the questioning of the right



to creation, which Mary Shelley sets in motion with her epigraph<sup>6</sup>, and therefore resonates with her own intentions.

The tragedy of *Frankenstein* is that by failing to comply with his creature's commands, and yet again denying him that which he wants most, Victor negates the only possibility for anything but a tragic outcome for both him and his creation. Consequently one should feel even more compassion for the creature, as he is dependent on Victor to change the course of events. Mary Shelley wants her audience to sympathise with the creature on the grounds that it is inherently good and is only driven to monstrous acts by the completely callous rejection and denial by the one man who has it in his power to bestow happiness on him. Furthermore, the co-dependency between the creator and the creation, which Mary Shelley sought to emphasise as Mellor points out, by using "wretch" to describe them both, is actively inverted by Percy Shelley's replacement of "devil", a word far less sympathetic, which emphasises yet again the threat of the creature rather than his disconsolate state. Percy Shelley's interference also terms the creature "an abortion", much in the same manner as he termed him a "devil", and keeping it in the same vein as his imposition that it sought revenge. Abortion is a powerful term to use as a description, which elaborates on the use of "devil" by pointing rather to the malformed, inherently evil nature of the creature. This also leads to the further absolution of Victor by the implication that it is the creature that is defective and not Victor, even though he is responsible for bringing him into being against the laws of nature. Percy Shelley's continual identification with Victor, his consistent reading of Victor as a sympathetic character, undermines much of what is significant about the text, but as the extract quoted above shows, it is not only the narrative of the creature which preserves Mary Shelley's intentions. The imposition of words such as "devil" and "abortion" to describe the creature as monstrous does not eliminate the integral elements within the text that point towards Victor Frankenstein's failure and above all, his having "no right" to withhold happiness from his creation. However, even when he is not directly editing the text, Percy Shelley's understanding of Victor never wavers. Mellor argues that:

As his review of the novel concludes, Frankenstein was not a perpetrator but only "the victim" of evil. Throughout the original text, Mary Shelley stressed Frankenstein's capacity for self-deception, while Percy, sometimes as blind as Frankenstein himself, softened or eliminated his errors. [...] He thus undercut her otherwise consistent portrayal of Frankenstein as an egotist who perceives only his own feelings and dangers. (Mellor 1989, 63)

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<sup>6</sup> As discussed in the previous chapter.

This shift from the original concept of Victor as perpetrator, and therefore as fallible even though his entire motivation for creation is to transcend his own fallibility, to “*only* the victim of evil” dulls the critique inherent in the novel. Mary Shelley stressed Victor’s egotism, and Percy Shelley “blind as Frankenstein himself” tried to eliminate these elements, almost as if to excuse the man who was so closely modelled on himself.

How Percy Shelley saw himself as a poet explains much of his unquestioning allegiance to Victor Frankenstein. Percy Shelley’s views of himself are remarkably similar to those held by Victor, who clearly states that his motivation for his creativity is so that future generations would laud him for he would “pour a torrent of light into our dark world. A new species would bless me as its creator and source; many happy and excellent natures would owe their being to me.” (M. Shelley, 36). Percy Shelley, in a letter to Hunt in 1816, the year in which Mary Shelley started working on *Frankenstein*, states that:

I am undeceived in the belief that I have powers deeply to interest, or substantially improve mankind. How far my conduct and my opinions have rendered the zeal and ardour with which I have engaged in the attempt, I know not [...] Perhaps I should have shrunk from persisting in the task which I have undertaken in early life, of opposing myself, in these evil times and among these evil tongues. (In Holmes, 351)

In these lines, Percy makes a confession about his character, which resonates hauntingly with Victor’s own view of himself. He believes that he can, through the pursuit of his creativity “substantially improve mankind” but more than that he sets himself up against as “opposing” to the “evil” times and tongues, implying perhaps a world of darkness in which he alone is the light. Therefore, by his own estimation Percy Shelley seems to think that through his life and the light that he epitomises; ultimately, he brings enlightenment to the world. Shelley sees himself as a reformer of the dark world, and persecuted by those that do not understand his light. The rest of the letter serves to further illustrate how misunderstood Percy felt himself to be, and how undervalued. It therefore comes as no surprise that perhaps in Victor’s character Percy Shelley felt himself looking at someone who he could identify with. Thus through his editing skills Percy Shelley gives Victor the support of a kindred spirit, the support of someone who understands what it means to be persecuted. Consequently, and with the flourish of a pen, Percy Shelley tries to obliterate the implied persecution or judgement on Victor’s character in Mary Shelley’s original text, becoming yet again the Romantic hero he always strove to be.

The identification that Percy Shelley actively inscribes into the original manuscript of *Frankenstein*, where he identifies sympathetically with Victor, enhances what is already a solid biographical connection between Percy and Victor. The critique of Victor's character, and consequently her "blistering critique" of the Romantic ethos, survive Percy Shelley's active interference in Mary Shelley's original text. The narrative told by the creature, on closer reading gives one a deeper insight into what Mary Shelley might have wanted readers to see as lacking within her protagonist. However, one of the major changes made by Percy Shelley within the line of associating himself with Victor in the face of a monstrous creation is solidified in his changes to the ending of the novel, as Mellor highlights:

More important, Percy changed the last line of the novel in a way that potentially alters its meaning. Mary penned Walton's final vision of the creature thus: "He sprung from the cabin window as he said this upon an ice raft that lay close to the vessel and pushing himself off, he was carried away by the waves, and I soon lost sight of him in the darkness and distance." Mary's version, by suggesting that Walton has only lost "sight of" the creature, preserves the possibility that the creature may still be alive; a threatening reminder of the potential danger released when men egotistically transgress nature and "read" the unknown as evil. Percy's revision, by flatly asserting that the creature was "lost in darkness and distance" provides a comforting reassurance to the reader that the creature is gone into the darkness and distance. We might go so far as to say that Percy's reading of the novel's conclusion is a defensive manoeuvre to ward off anxiety and assert final authorial control over his wife's subversive creation. (Mellor 1989, 68)

The ending as it now survives, is Percy Shelley's final say on the creature's fate. In the 1818 text, he does not push himself off from the vessel thereby letting the waves move him into the distance but,

He sprung from the cabin-window, as he said this, upon the ice-raft which lay close to the vessel. He was soon borne away by the waves, and lost in darkness and distance. (M. Shelley, 191)

The original unaltered version, which Mellor cites, maintains the creature's agency that leaves his fate more under his control than that of the icy floes of the pole. By negating the creature's agency in relation to his own fate, it does indeed seem as if this tiny change to the original text has larger ramifications than merely shortening the sentences. By making the creature an active agent, in the original version, Mary Shelley does preserve not only "the possibility that the creature may still be alive" but also, and perhaps more importantly, that the creature retains a strain of humanity at the ending of the story.

Throughout the novel, there is a tension between the creature's version of himself and how he is perceived by Victor and by association Percy Shelley. The creature states repeatedly that he is inherently good, but has been rejected by his creator and ostracised by society. His complete abandonment by the world is what makes him essentially one of the loneliest characters in literature. Victor, on the other hand, is continually unwilling to see the human element in his creation, as he is consistently horrified by the inhuman physical qualities which he has created and cannot look beyond that to acknowledge the desire of his creation for companionship. Mary Shelley uses the ugly physicality of the creature for a dual purpose. Firstly, the reaction of society when confronted with the creature highlights the tendency of people to feel threatened by what they see as ugly or horrifying, specifically in the late eighteenth century context where people generally relied on their belief that facial structure indicated personality and general character traits. Secondly, Mary Shelley is using Victor's reaction to his own creation to comment on his general inability to deal with the reality of the outer world. Victor as a Romantic is unable to deal with something real that falls outside of his drive for transcendence or attaining that transcendence. Also, the ugliness of the creature is a consistent reminder of Victor's failure; self-involved as he is, he never thought about the reality of what he was creating, but saw only the act of creation in itself as important. This tension creates a binary between Victor's aversion to his creation, and the creature's human need for society. Repeatedly in the novel, it is highlighted how the creature is misjudged by virtue of his appearance alone, something which is Victor's direct fault, but which never actually occurs to him. Victor, shuddering at the sight of what he has created, bursts out:

'You have left me no power to consider whether I am just to you or not. Begone! Relieve me from the sight of your detested form.' 'Thus I relieve thee, my creator,' he said and placed his hated hands before my eyes, which I flung from me with violence. (M. Shelley, 79)

Victor's inability to acknowledge the humanity of his creation is heart-wrenchingly apparent in this extract. He flings away the pleading hands, which relieve him of the hated sight of the creature who still speaks to him as "my creator", with violence. It is in this manner that the creature is ultimately flung from the narrative at the very end.

One could almost say that the unfeeling casting-off of the creature's hands by Victor is mirrored by Percy Shelley casting-off the creature in a similar fashion at the end of the novel. Percy Shelley's imposed and abrupt ending where the creature disappears without agency, without relief, stands in stark contrast with the creature's last words, which Percy Shelley has left

unchanged from the original manuscript. The almost completely opposite tones of the ending and the creature's last words are emphasised further if one takes into account that in the creature's last speech act, there is an inevitable emotional response from the reader that is elicited in much the same way as his previous addresses evoke sympathy by consistently highlighting his humanness. Yet this one is perhaps the most significant of all, as he addresses Victor Frankenstein, even though dead:

'Farewell! I leave you, and in you the last of human kind whom these eyes will ever behold. Farewell Frankenstein! If thou wert yet alive, and yet cherished a desire of revenge against me, it would be better satiated in my life than in my destruction. But it was not so; thou didst seek my extinction, that I might not cause greater wretchedness; and if yet, in some mode unknown to me, thou hast not yet ceased to think and feel, thou desirest not my life for my own misery. Blasted as thou wert, my agony was still superior to thine; for the bitter sting of remorse may not cease to rankle in my wounds until death shall close them forever:

'But soon,' he cried with sad and solemn enthusiasm, 'I shall die, and what I now feel be no longer felt. Soon these burning miseries will be extinct. I shall ascend my funeral pile triumphantly, and exult in the agony of the torturing flames. The light of that conflagration will fade away; my ashes will be swept into the sea by the winds. My spirit will sleep in peace; or if it thinks, it will not surely think thus. Farewell.' (M. Shelley, 191)

In this rather lengthy yet poetic final soliloquy, we find, not a monstrous creature exulting in the death of a creator who spurned him, abandoned him and broke every promise he ever made, but rather a saddened creature, who with every turn of his phrases, emphasises his own humanity. In his last speech act, the creature is obviously distressed and stresses how he was pained by this opposition with Victor and how "my agony was still superior to thine." He acknowledges Victor's pain, but points out that Victor has wreaked his revenge rather "in my life than in my destruction", and that this pain, along with the remorse that he now feels "may not cease to rankle in my wounds until death shall close them forever". The creature is tormented by the tragic turn of this tale, rather than monstrously joyful at the demise of his opponent, as Victor's version of him had expected him to be. Mary Shelley emphasises his pain by painting it viscerally and dramatically in using phrases such as "bitter sting", "burning miseries" and ashes being "swept into the sea", which are all effective in the human empathy they elicit for this tortured creature who even refers to himself as having a "spirit" which "will sleep in peace". There is also a certain dignity to his last speech act, in his promise to "ascend my funeral pile triumphantly and exult in the agony of the torturing flames". Completely alone now, without even his creator, the creature is aware that he will not survive long, but is acknowledging the inevitability of his death, however painful it might be, with an almost calm dignity. This dignity, sadness and the solemn nature of the creature's farewell is closely linked to Mary Shelley's continual empathy for him.

The last two lines of the novel are therefore all the more at odds with this last empathetic insight into the creature's inner being, which proves that he is in fact not monstrous at all. The last two sentences, where he is flung seemingly carelessly into the distance and darkness, is reminiscent of Percy Shelley's abrupt misreading and editing. Pushing himself off, letting the ice raft drift him away, as the original intended would have lent a certain dignity to the creature's exit, implying perhaps that he has now, now that there is not a single person left to sympathise with his cause, chosen his own isolation and exile, unwillingly but graciously.

The fact that the reader is not given certainty of the creature's death in either version is an important point to focus on as it can be read in various ways. Mellor seems to see this lingering presence of the creature in Mary Shelley's original manuscript serving as "a threatening reminder of the potential danger released when men egotistically transgress nature and 'read' the unknown as evil." (Mellor 1989, 68) As there seems to be a cautionary element to the novel at large, this is one way of interpreting it. However, this "threatening reminder" is also tempered by the creature's last speech, which arguably places the emphasis rather on the danger of men "egotistically" reading the unknown as evil rather than the creature as "potential danger released". The abrupt ending of the novel perhaps enhances the "potential danger" Mellor is here referring to, and therefore is another testament to Percy Shelley misinterpreting *Frankenstein*. Aligning himself fully with the "modern Prometheus" Percy Shelley coldly ends the creature's lingering presence, seemingly untouched by his final words of farewell.

The link between Victor Frankenstein, Percy Shelley and the notion of the "modern Prometheus" is further paralleled by the fact that Victor Frankenstein practises the same editorial control over Walton's journal account of his experiences as Percy Shelley practises over Mary Shelley's manuscript:

Frankenstein discovered that I made notes concerning his history: he asked to see them and then himself corrected and augmented them in many places; but principally in giving the life and spirit to the conversations he held with his enemy 'Since you have preserved my narration,' said he, 'I would not that a mutilated one should go down to posterity. (M. Shelley, 179)

Victor therefore becomes an author in the same sense that Percy Shelley actively interfering with Mary Shelley's text seems to imply that he has changed the slant of the text. What is particularly interesting about this extract, apart from setting up another similarity between Victor and Percy,

is Mary Shelley's choice of the word "mutilated" to describe an unedited text. It is almost as if there is an anxiety of becoming an author which has found vent in this extract, which links to her description of her novel as her "hideous progeny",<sup>7</sup> or perhaps even her irritation at the text unedited being deemed mutilated or broken by the person who is editing it. Victor Frankenstein being linked, however subtly, to an author, is important in this instance not because of this link to Mary Shelley, but rather in regards to how this associates him more substantially with another contemporary Romantic figure, namely the cursed Mariner who is also the teller of his own cautionary tale. This connection further enhances the haunting echoes of *The Rime of the Ancient Mariner* that are present in the text due to Walton's character, as has been discussed earlier, by adding Victor as another association to this epic poem. Especially the latter aspects of the plot within *Frankenstein* are reminiscent of the Ancient Mariner being pursued by a shadowed fate born out of his own transgression against nature. Ultimately, Frankenstein, like the Ancient Mariner, finds that he is alone on the wide expanse of the ocean, terrified and alone, and at the sight of land, he cries in relief:

Almost spent, as I was, by fatigue, and the dreadful suspense I endured for several hours, this sudden certainty of life rushed like a flood of warm joy to my heart, and tears gushed from my eyes. (M. Shelley, 144)

This seems to be a close paraphrase of Coleridge's own words in *The Rime of the Ancient Mariner*, the Mariner, upon seeing a "sudden certainty of life" represented by the water snakes exclaims: "A spring of love gushed from my heart, / And I blessed them unaware" (lines 284-5). Interestingly, Mary Shelley also makes use of another of Coleridge's poems to explain Victor's state. In *Frost at Midnight* when Coleridge explains the image of the poet's mind as that "which fluttered on the grate/ still flutters there, the sole unquiet thing" (lines 15-6). This "sole unquiet thing" is similar to the words used in Mary Shelley's definition of Victor Frankenstein's state of mind in contrast with the stillness of the night surrounding him:

I was often tempted, when all was at peace around me, and I the only unquiet thing that wandered restless in a scene so beautiful and heavenly, if I except some bat, or the frogs, whose harsh and interrupted croaking was heard only when I approached the shore – often I say, I was tempted to plunge into the silent lake, that the waters might close over me and my calamities forever. (M. Shelley, 70)

With the imagery then as well as consistently paraphrasing Coleridge, this particular aspect of *Frankenstein* cannot be ignored.

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<sup>7</sup> Mary Shelley, *Frankenstein or The Modern Prometheus*, London, Penguin Books(2003), pg 10

The most important question to answer is why Mary Shelley, who is constantly undermining Romanticism, would allow an important Romantic's images and phrases to appear in whatever form within her novel. She consistently subverts Romanticism throughout, and yet the resonances of the Coleridgean elements within her novel seem to imply a conflict. Based on this inclusion it would be an oversimplification merely to assume that *Frankenstein* being infused with certain aspects of Coleridge's poetry destabilises her questioning of Romantic ideology. In fact, quite the opposite is true. It is precisely the inclusion of such paraphrases that maintains *Frankenstein* as a Romantic text that simultaneously questions Romanticism. It thereby becomes a Romantic text about Romanticism, making it self-reflexive or in other words, a self-conscious attempt to show those aspects within Romanticism that are particularly threatening or dangerous from within that ideology. By consistently paraphrasing a leading poet of her time, Mary Shelley is also intertextually relating Victor Frankenstein as a scientist to the figure of a poet. She is infusing the words from Coleridge into the minds and utterances of both Walton and Victor, aligning them therefore specifically with the literary Romantics. Reading the resonance in this way would mean that through intertextual references of Coleridge writings, Mary Shelley clarifies, furthers and amplifies her critique of Romantic thought and its egotistical ethos to include Romantic poetry or the poet inclined to Romanticism. Therefore by including Coleridge's utterances specifically in Victor's thoughts or mind she is criticising the ideas in his poetry as being similar to Victor as a Promethean figure. Even though Coleridge often shows the alienation implicit in the Romantic notion of solitude, such as his figure of the Ancient Mariner, the implication for Mary Shelley is that he is not fully developing that notion. Her use of Coleridge, who was aware of the alienating element of Romanticism, implies that she was aware that not all Romantic poets are fanatical or extreme in their adherence to an absolute and idealised Romanticism. However, creating a parallel between Victor and the Mariner suggests rather that to some extent all Romantic poets deal with or create Promethean archetypes in their creative work. She acknowledges Coleridge's articulation of alienation but points to the fact that his Mariner remains in many ways a Promethean figure, albeit a darker and slightly distorted one. Consequently one could argue that Mary Shelley questions the manner in which Coleridge deals with the Romantic aspects in *The Rime of the Ancient Mariner*, while simultaneously using his work to enhance the Romantic elements in Victor's temperament and make up. This is perhaps why there are so many parallels between Victor and the Ancient Mariner, transgressors who are punished but only survive to tell their cautionary tale.



It is in the difference between the Ancient Mariner and Victor that her criticism becomes apparent. Victor diverges from the Ancient Mariner in that his estrangement from society is more consciously sought and the ramifications of his estrangement are more severe. The Mariner is in some ways still able to redeem himself by becoming a bringer of wisdom to others, whereas Victor does not absolve himself and his attempt to do so fails utterly. The reality of *Frankenstein* is fundamentally more powerful than in *The Rime of the Ancient Mariner* as the Ancient Mariner redeems himself towards the end of the poem as he states that “I pass, like night, from land to land; / I have strange power of speech;” (lines 586-7). The Ancient Mariner has a “power of speech” that by implication transcends the mortal realm as he is asked “What manner of man art thou?” (line 577). Therefore, the Ancient Mariner becomes a bringer of wisdom with his tale and attains at least in part the Romantic ambition towards transcendence. He is alone, “wrenched / with a woeful agony” (lines 578-9), but he is never fully human again, thereby attaining in a sense a divinity that Mary Shelley deliberately denies Victor, who unlike the Ancient Mariner does not see his own fault, and dies failing to attain any of the Romantic goals he has set himself. Thus, her use of Coleridge’s work underlines the Romantic connection of *Frankenstein* while at the same time drawing attention to where her judgement of Victor is more severe. Again, this strengthens as well as expanding her focus. It allows her criticism to apply to the ideology of Romanticism itself rather than simply commenting on Percy Shelley through Victor Frankenstein. Her criticism extends to Romantic presumption or at least to the Romantic’s insistence in perpetually dealing with the Promethean paradigm. However, even with her use of Coleridge’s interpretation of an alienated transgressor, Percy Shelley remains among the first and foremost of those Romantics that her novel criticises.

Percy Shelley encouraged Mary Shelley to write her novel, because he himself believed that writing was a way in which to lead society to revolution, to create a new truth or reality, to bring light into the dark world. His Romantic worldview seems to have been undiluted to the point where his focus on reform starts to resemble Victor’s drive to create, to alter the world for the better at the exclusion of everything else. Percy Shelley was passionate about altering the world through poetry to the point of declaring that “Poets are the unacknowledged legislators of the world” (in Hoobler, 66). This exclusion would then explain how he could misread *Frankenstein*, but Percy Shelley’s *Prometheus Unbound* implies that perhaps he was not as completely ignorant of Mary Shelley’s criticism as his editing indicates. As Badalamenti indicates, there is evidence of his

unconscious perception of his own involvement in the creation of the character of Victor as the modern Prometheus:

He (Percy) began writing *Prometheus Unbound* in the autumn of 1818, about one and a half years after Mary finished *Frankenstein*. He finished it around April of the next year. It represents a liberation of the human spirit in a number of ways and the imagery of the title suggests a longing for freedom. If *Frankenstein* stands for Percy as an irresponsible lover/spouse then *Prometheus Unbound* makes his reply to Mary's critique to be exempt from its claims. (Badalamenti, 433)

One cannot ignore the fact that Percy Shelley is in some way establishing a dialogue with Mary Shelley's text beyond his active interference with her original manuscript of *Frankenstein*. Therefore, the notion that *Prometheus Unbound* is a justification of Promethean ambition, and therefore a reaction to Mary Shelley's criticism of such ambition, is a valid one. This is particularly true when one takes into account Percy Shelley's introduction to his poem, where he unequivocally states in his description of Prometheus that:

The only imaginary being resembling in any degree Prometheus, is Satan; and Prometheus is, in my judgement, a more poetical character than Satan because, in addition to courage and majesty and firm and patient opposition to omnipotent force, he is susceptible to being described as exempt from the taints of ambition, envy, revenge, and a desire for personal aggrandisement [...] Prometheus is, as it were, the type of the highest perfection of moral and intellectual nature, impelled by the purest and truest motives to the best and noblest ends. (P. Shelley, 776)

In this explanation of the Promethean character, one again gains insight into why Percy identified almost without hesitation with Victor Frankenstein's character. As the "modern" Prometheus, Victor Frankenstein is therefore a continuation of what Percy Shelley esteemed as "the highest perfection of moral and intellectual nature" that consequently could only be driven by what he terms "the purest and truest motives" to attain "the best and noblest ends". Percy Shelley then reinstates the inspirational aspect of the Promethean character that the Romantics were so fond of aspiring to. The fact that he uses the words "highest", "best", "noblest", "truest" and "purest" in one sentence to explain the most important character traits of his Prometheus, is excessively idealised prose, indicating repetitively how Prometheus has no faults. This is precisely what Mary Shelley did not see in her rendering of the Promethean myth. Even though Victor, as the modern Prometheus, wants to attain transcendence over human character traits, he cultivates and exhibits character traits that could not be described with any of the words that Percy Shelley uses to plot his expectation of a Promethean character. Victor Frankenstein is even more human precisely because of his complete failure and it is here that the strength of Mary Shelley's questioning of the idealised Prometheus is at its most commanding. Throughout her plot and the character development within *Frankenstein* Mary Shelley shows remarkable

insight into the reality of such high-flown motivations, presenting to the reader how easily someone who feels it is their duty to enlighten a darkened world overreaches to a point where the only possible outcome is a tragic one. If one accepts that these different opinions about Prometheus could be seen occurring almost as if in dialogue, the difference of opinion becomes even more apparent. On the one hand, one has *Frankenstein*, Mary Shelley's text and her insightful grounding of the myth into a flawed reality where it is the flaws of idealisation and the presumption of Romanticism that are illustrated. On the other, one has Percy Shelley's editing of her original manuscript and then his unwavering praise of Prometheus in his poem *Prometheus Unbound*. Consequently, Mary Shelley and Percy Shelley seem to be continually in opposition within this argument about Romantic ideology.

The difference between Mary Shelley's and Percy Shelley's views on Prometheus hinges on the fact that Mary Shelley did not fully subscribe to the overly idealistic points of Romanticism. Instead of being drawn into a glorification of these Romantic ideals, Mary Shelley sets herself apart by instilling a sense of realism into the story. However, it would detract from the complexity of her work if one did not acknowledge that even her criticism of these ideals is not in fact a complete rejection of Romanticism as a whole. As *Frankenstein* remains in many ways a Romantic text, it takes on the form of an insightful cautionary tale in which the realistic or rational implications of Romanticism are examined rather than condemning Romanticism completely. Mary Shelley writes her Romantic idealist into a world where isolation from society does not bring inspiration that benefits all of humanity. Rather this isolation leads to alienation with tragic consequences, where nature is terrifying as often as it is calm or sublime, where acquiring knowledge in itself is unpunished, but actively implementing that knowledge without the thought of the consequences is rewarded with death. There is a pattern established here which is consistent in all the aspects that Mary Shelley questions or undermines within Romanticism, namely that she permeates the egotistical overreaching presumptions with realistic and tragic consequences.

However, what makes *Frankenstein* all the more noteworthy is that, regardless of Percy Shelley's impositions the integrity of her criticism remains intact. The intent of Mary Shelley to question and undermine Romanticism can only be appreciated fully if the reader does not become ensnared as Percy Shelley and Victor are, by disregarding the creature either as unimportant or

monstrous. It is exactly this oversight and over-identification with the protagonist on Percy Shelley's part which most strongly seems to prove Mary Shelley's point. One could argue therefore, that this is why in spite of these impositions *Frankenstein* maintains all the elements that Mary Shelley intended. Interestingly, if one keeps in mind that it is precisely in the creature's narrative that Mary Shelley's voice remains unaltered and it is at this point that her criticism of Romanticism is at its most unchanged, then one can also utilise the creature's narrative to show that Mary Shelley was not polarising her opinion to being anti-Romantic. In fact, Victor's creation never wanted to be regarded in opposition to Victor. There is a fundamental need in the creature to be accepted specifically by the one who has made him to be what he is. Throughout the creature's final speech, there is a definite sadness as he addresses Victor because "thou didst seek my extinction" (M. Shelley, 191). Throughout the creature's narrative there is a yearning for reconciliation, for Victor to change his ways so as to include him or at least take responsibility for his existence. Mary Shelley draws attention to all the darkness and unhappiness brought about by the pursuit of Romantic ideals with dramatic consistency as they drive Victor towards his death. However, as a cautionary tale, *Frankenstein* highlights not only the dangers of Romantic ideology, but also points the reader towards an alternate path. It becomes increasingly clear that if Victor had only ceased in his solipsistic drive towards transcendence and self-deification, the tragic events of *Frankenstein* might have turned out very differently.

Consequently, the criticism of Percy Shelley that exists within Mary Shelley's text is not an absolute or completely damning criticism, in the same way as she does not condemn Romanticism in its entirety. The creature is at times angry and enraged, hurt, abandoned and rejected, but he never abandons Victor, not even in death. Moreover, if it is through this character that Mary Shelley was articulating her own response to Romanticism then the response is not merely an outright criticism, but a nuanced response where the prevailing emotion specifically at the end of the novel, is one of sadness. This sadness permeates the ending because the creature makes clear that his own feelings of loss and anguish are a result of Victor's death, and that if Victor had only been able to acknowledge his existence as positive or taken responsibility for that existence, then the conflict between them could have been resolved. A nuanced response to Romanticism also allows for the relationship that existed between Mary Shelley and Percy Shelley to have been what it was, as Mary Shelley allowed Percy Shelley's impositions into her text. No matter how he misunderstood her intentions, she allowed him to edit, which is only perplexing if one sees her criticism as a total condemnation of his character. It

is precisely for this reason perhaps that Percy Shelley's editing, which at times is completely insensitive towards or ignorant of this nuance within the text, is baffling. Perhaps his editing of her text could have been a redemptive act for him by giving him the opportunity to acknowledge the validity of something that falls outside of his own worldview, and like Victor Frankenstein, he fails to allow himself that redemption. As his editing continually shows, he in many ways doggedly reinscribes Victor as a victim, a hero and refuses to see Victor's creation as anything but a "devil" or an "abortion", thereby dooming Victor as well as himself to being read as precisely the type of egotistical Romantic persona that Mary Shelley was trying to warn against.

University of Cape Town

### Chapter 3: The monster redefining boundaries between genres

And then I dived,  
In my lone wandering, to the caves of death,  
Searching its cause in its effect; and drew  
From wither'd bones, and skulls and heap'd up dust,  
Conclusions most forbidden. ~ Byron, *Manfred*, (II, 79-83), (1817)

As shown in the previous chapters' discussions of *Frankenstein*, Mary Shelley skilfully navigates her criticism of idealised Romanticism through her personal experience, particularly of Percy Shelley. Sceptical and consistently critical of Romantic egotism, Mary Shelley uses Victor to highlight precisely how dangerous a pursuit of Romantic principles can be. Formulating her interrogation of Romanticism from within Romanticism itself makes *Frankenstein* especially interesting in relation to literary genre categories or modes. Drawing on her contemporary notion of Romanticism and literalising it through Victor, Mary Shelley questions certain aspects of Romanticism while simultaneously bringing them into play. This ultimately means that *Frankenstein*, as a literary text, is both of the Romantic tradition and outside of it, crossing the boundaries between Romanticism and other, which is why it overlaps or permeates into what is perceived to be the Gothic tradition. On defining the Gothic, it becomes clear that the boundary between Romanticism and Gothic is a tenuous one as DeLamotte suggests:

Because the question of the distinction between the me and the not-me is central to light as well as "dark" Romanticism, the definition of Gothicism as fundamentally concerned with the boundaries of the self provides another way of looking at the connection between the Gothic tradition and the Romantic tradition. (DeLamotte, 23)

By using the terms "light" and "dark" Romanticism to make a distinction between Romanticism proper and the Gothic, DeLamotte highlights the fact that both genres are at their cores, still originating from the same nexus and concerned with very similar questions of the self in relation to society. This mutual concern is termed by DeLamotte as "the distinction between the me and the not-me", or in other terms, the boundary between the self and the other. The text of *Frankenstein*, then, much like a Gothic protagonist, transgresses a boundary, specifically the one that would lie between the two genres, and simultaneously inhabits both. What makes DeLamotte's use of the terms more interesting is that one can arguably then investigate this relationship between the Gothic tradition and the Romantic tradition, and see how they are connected, and in this case, with specific reference to Mary Shelley's text, as the issue of the monstrous nature of Romanticism unbound is central to understanding *Frankenstein*. Extrapolating from this link between the two genres, it seems as if there is a continual

connection or preoccupation, however tenuous, which links them. This almost necessitates that *Frankenstein*, by criticising Romanticism as it does, be a Gothic text, but as I will show later in this chapter, this is not exclusively true.

As Hume suggests this connection is far from being a new discovery; he illustrates in *Gothic versus Romantic* that Gothic writers were very much aware that they were writing “as part of a resurgence of romance against neoclassical restrictions” (Hume, 282). The shift towards Romanticism is explored further within the Gothic literature of the time as another “symptom” of moving away from what Hume calls “neoclassical ideals of order and reason”, and reaching towards highlighting the Romantic ideals of experiencing emotion and most importantly, a focus on the imagination. The problem for Hume is expressed not in the relation between the genres, but in how that relation has been perceived in literary thinking:

That Gothicism is closely related to romanticism is perfectly clear but it is easier to state the fact than to prove it tidily and convincingly. There is a persistent suspicion that Gothicism is a poor and probably illegitimate relation of romanticism, and a consequent tendency to treat it that way. There are those, indeed, who would like to deny the relationship altogether. (Hume, 282)

Even though the capitalisation of “Gothicism” versus the smaller “romanticism” in this extract and throughout Hume’s argument clearly shows where his sympathies lie, it is important to pause over what he is saying and acknowledge that this difficulty of “proof” is in fact true. For the sake of this argument, therefore, the trajectory of trying to highlight precisely how intimately Romanticism and Gothicism are related in and through *Frankenstein*, is not at the outset trying to prove that they are as ultimately intertwined as genres, but rather to examine how Mary Shelley uses aspects of both genres to establish within her first novel, something new. Clery demonstrates a subtle understanding of this interplay between established genres and the advent of something original in Shelley’s text:

The Preface to the first edition of *Frankenstein* aims to distance the novel from hackneyed Gothic convention, but in doing so it re-emphasises one element which, it has been argued here most crucially characterises the genre. We are told that the “event on which this fiction is founded” – the creation of a man from inanimate matter - has been considered by scientific writers “as not of impossible occurrence”. That is what distinguishes this story, however incredible, from “a mere tale of spectres of enchantment”. (Clery, 117)

As Shelley’s critique of Romanticism distances her text from becoming a complete Romantic text, so, as Clery states, in the preface to *Frankenstein*, Shelley is actively trying to distance her

novel from Gothicism. This is achieved by ascribing the happenings of the plot “as not of impossible occurrence”, which in relation to Gothic writing is a step away from the intervention of the supernatural or “tale of spectres of enchantment”, which is therefore of impossible occurrence, that is used by Gothic writers to induce both horror and terror in the reader. Mary Shelley’s distancing, as Clery points out, is to attempt something outside of the “hackneyed Gothic convention” which at the same time re-establishes the Gothic, but as in the case of Romanticism within her text it underlines and undermines those concepts that she is trying to move away from. It is therefore quite correct to assume that “like all innovative successes *Frankenstein* was both deeply familiar to its original audience, and shockingly new” (Clery, 127). It is precisely this navigation between the two genres that is both remarkable and of particular interest, and it is to this interplay that one must pay exceptional attention. Shelley redefines and re-establishes, conflates and mutates almost all aspects of both Romanticism and the Gothic to create her “hideous progeny”, in much the same way as Victor does in the creation of his “monster”, through piecing together fragmented and often strangely opposing elements, from the enlightened knowledge of his science and pieces of dead flesh from the graveyard.

If one looks more closely at the “dark” side of Romanticism, it becomes clear that the transgression or anxiety of transgression that so pervades it is, as Botting suggests, necessary to a more complete contemplation of what he terms as modernity:

[Gothic] shadows the despairing ecstasies of Romantic idealism and individualism and the uncanny dualities of Victorian realism and decadence. [...] In the twentieth century, in diverse and ambiguous ways, Gothic figures have continued to shadow the progress of modernity with counter-narratives displaying the underside of enlightenment and humanist values. Gothic condenses the many perceived threats to these values, threats associated with supernatural and natural forces, imaginative excesses and delusions, religious and human evil, social transgression, mental disintegration and spiritual corruption. (Botting 1996, 1-2)

This shadowing, which is yet again a reiteration of the dark side of the light metaphor, is in this extract closely linked to being a counter-narrative to the “progress of modernity”. The use of the word “modern” in relation to *Frankenstein* is also underlined by the subtitle of the text, “*The modern Prometheus*”, establishing a very clear link between the text and a modern incorporation of an older form or myth. This extract also sets up Gothic writing as a reactionary literary form, and which is arguably why it seems porous, unstable, and therefore as mentioned by Hume as being “a poor and probably illegitimate relation of romanticism”. Botting then continues by listing prototypical Gothic villains and interestingly incorporates the notion that



This list grew, in the nineteenth century, with the addition of scientists, fathers, husbands, madmen, criminals and the monstrous double signifying duplicity and evil nature. (Botting 1996, 2)

Due to the fluidity of Gothic writing, there seem to be continually changing, albeit desolate and menacing, landscapes of possibilities to articulating that anxiety about the “progress of modernity”. This progress is what leads ultimately, in the mind of DeLamotte, to there being aspects which recur within Gothic writing, namely that there are “two fears [that] dominate this Gothic world, the fear of terrible separateness and the fear of unity with some terrible other.” (DeLamotte, 22) For DeLamotte these uncertainties or anxieties are based around boundaries and their transgression, which are highlighted when the protagonist is confronted with violence:

The psychological, moral, spiritual and intellectual energies expended in the engagement with the forces of violence, are generated by an anxiety about boundaries: those that shut the protagonist off from the world, those that shut the protagonist in and those that separate the individual self from something that is other. (DeLamotte, 19)

As a Romantic idealist, Victor continually wants to transcend boundaries and limitations, so there is a sense in which his Romantic drive is in part an “anxiety about boundaries”. However, this drive towards transcendence in Romanticism is translated as being a transgression within the Gothic. In undermining Victor’s need for transcending human limitations, Mary Shelley continues to highlight the boundaries that Victor cannot break through. As a result of Victor being human and fallible, he fails to attain Promethean status, which leaves him contained within the boundaries that DeLamotte lists in her extract. Not only does Victor actively shut himself “off from the world”:

And the same feelings which made me neglect the scenes around me caused me also to forget those friends who were so many miles absent, and whom I had not seen for so long a time. (M. Shelley, 37),

but outside of this, he is also “shut in” “in a solitary chamber, or rather cell, at the top of the house [...] my workshop of filthy creation” (M. Shelley, 36). The boundary that separates Victor’s individual self from something that is other, namely that which he has created, is the horror that he feels every time he looks upon his creature, effectively keeping himself from reaching out to his own “other”. The boundary that Victor is unable or refuses to transcend is the one between himself and his creation. It is this boundary, as Mary Shelley’s criticism implies, that should be breached rather than the original boundary Victor breaks by creating life out of death. It is precisely his presumption of becoming a creator that makes Victor the “modern Prometheus” and again links him both to Romanticism and the Gothic:

The titanic hero – villain, or heroine – villainess of many Gothic works is fashioned after Prometheus or Faust, archetypal transgressors of the dividing line between the human and the divine. (DeLamotte, 22)

Victor becomes this “archetypal transgressor” right from the beginning of *Frankenstein* by perceiving life and death “as ideal bounds, which I should first break” (M. Shelley, 36). The Romantic notion of defining the self as solitary yet exalted in its search for creative or imaginary truth in the same vein as Prometheus, is undermined here by a reference to the Gothic convention of anxiety about boundaries. Mary Shelley roots her Romantic hero, who wishes to go beyond all limitations, firmly within a maze of anxiety-riddled boundaries, some breached and others insurmountable. Victor is enclosed in his “workshop of filthy creation”, and excludes himself not only from friends and family but also most importantly from his own creation. He oversteps the dividing line between the human and the divine, but fails to attain the divinity he seeks because Victor Frankenstein’s pursuit of knowledge is also flawed due to another Gothic flourish:

One of the problems of knowledge that Gothicists investigate is the dilemma of the self unable to perceive anything but its own reflection. (DeLamotte, 24)

In contrast to this dilemma, Romanticism celebrates the pursuit of knowledge, especially when it is a solitary genius reaching some form of transcendence or enlightenment, which will ultimately bring light to the world and render the solitary genius immortal. The Gothic problematises this linear course of action by highlighting the danger of falling into a cyclical trap, where the self that only sees itself drives his quest for knowledge or in other words, where only self-perception becomes possible. For the Gothic writer such a lust for knowledge is what seems to lead to tragedy, as there are some things, such as the creation of life in the case of Victor Frankenstein, that should not be known at all. For Mary Shelley, the boundary between acquiring knowledge and penetrating beyond the boundaries of life and death to pursue his thirst for knowledge is indeed an important one. It is only once Victor has isolated himself completely in his “solitary chamber or rather cell” and actively brings his knowledge into actual physical being that his deteriorating health should be read as an ominous prediction of the terror that his unfettered imagination and his lust for knowledge is about to unleash. His reaching for enlightenment is transformed into monstrous overreaching, reconfiguring Prometheus from a hero to a monster through Victor’s selfish need.

The notion of the sublime is another point at which the interests of Romanticism and the Gothic correspond, which is subtly demonstrated by *Frankenstein*. Romanticism saw the sublime as another departure from the classical technical integrity of literary work, shifting towards provoking ecstasy through the emotional and imaginative transcendence that surpassed all the classical notions of rules of style. This ability to transcend bounds, to achieve sublimity, was important to the Romantics, as it is one of the foundations of the entire movement, focusing specifically on the notion of exalting the imagination to perceive new truth. Edmund Burke in *Origin of Our Ideas of the Sublime and Beautiful* (1756) stated in relation to the sublime that:

Whatever is fitted in any sort to excite the ideas of pain and danger, that is to say, whatever is in any sort terrible, or is conversant about terrible subjects, or operates in a manner analogous to terror is a source of the “sublime”, that is, it is productive of the strongest emotion which the mind is capable of feeling. (Burke, 39)

This extract is important, as Burke illustrates by using phrases such as “pain and danger”, “terrible” and “terrible subjects” how this terror is a source of the sublime. This notion of the sublime is not found in the pastoral transcendence of Wordsworth, but is leaning more towards a Coleridgean understanding of the sublime. Coleridge’s poem *The Rime of the Ancient Mariner*, is of this school of eliciting the sublime, as Punter illustrates:

Most of those writers traditionally considered to be major romantic poets were influenced by, and played a part in shaping the evolution of the Gothic [...] When we turn to Samuel Taylor Coleridge, we find less emphasis on this political dimension and more on psychological mood, in the characteristically Coleridgean range of dejection, disappointment and melancholy. (Punter, 13-14)

The important distinction that Punter is highlighting is that even though Coleridge is by no means considered a Gothic poet, his focus was very different from his contemporaries because his focal point was not merely an idealised Romantic ideology, but more “psychological” in his focus on “dejection, disappointment and melancholy”. This “negative focus”, if one can call it that, is why *The Rime of the Ancient Mariner* contains a definite melancholic strain, an alienation that pervades the text rather than glorious solitude. Coleridge’s “negative focus” specifically in Gothic terms is therefore grounds to incorporate Coleridge’s work into *Frankenstein*, which falls outside of the Romantic considerations explored in the previous chapter. Mary Shelley diverges from Coleridge’s effect of the sublime, which he achieves partially through “psychological mood”, by using Burke’s sublime of “terror”. This sublime of “terror” merges the Gothic element of achieving the sublime through subversive means and the Romantic element where the sublime is a means of transcendence. This merging of the Romantic and the Gothic enhances both, as Botting illustrates:

Gothic, in adding, or returning, those elements of darkness and misfortune to the romance, injects something else, something different, into an evacuated form, renewing intensity and revitalising desire with objects and plots that seem more dangerous, real and credible; romance furnishing grand oppositions and limits to be transcended, allows gothic to glimpse as it wallows in its own superficially gloomy depths and sham figures, something more meaningful, elevating its night world to a reflection of higher human quests, hinting at something of the sacred. (Botting 2008, 22)

As this extract reveals, Botting believes that it is in the conflation or combination of Gothic and Romance forms that “something more meaningful” is explored. He describes Gothic as wallowing “in its own superficially gloomy depths and sham figures” whereas Romance is seen as an “evacuated form” that needs a renewal of both intensity and depth. It is in the combination of these elements that elevates the interaction of both “to a reflection of higher human quests, hinting at something of the sacred”. This is in fact exactly, what Mary Shelley achieves by combining both forms to a certain extent, highlighting her remarkable ability to supplement both forms with what is lacking. This is done by manoeuvring between the two, and simultaneously elevating the Gothic “night world” beyond that of the wallowing Gothic and elevating the Romantic element with depth by inducing a sense of reality to what easily becomes an optimistic yet shallow plot. Thornburg also examines how Mary Shelley intertwines the two forms more precisely through her characters:

Mary Shelley's *Frankenstein* is in one sense an antisentimental novel; that is, it is a novel in which the sentimental tradition is consciously invoked so that its flaws may be ironically revealed [...] *Frankenstein* is covertly antisentimental; the characters with whom the reader is invited to sympathise are themselves determinedly sentimental from start to finish. Victor Frankenstein, who is both instrument and victim of the book's irony, is aware of the irony only imperfectly. The only character who actually comes to a perception of the sentimental tradition's flawed nature is the Monster, the novel's apparent antagonist, with whom the reader is not free to sympathise openly. Thus *Frankenstein's* antisentimental nature is withheld, bound up with the book's Gothic, mirror image truths in a way that arouses and maintains the reader's anxiety but never quite dispels it. (Thornburg, 63-64)

In this extract Thornburg draws attention to the fact that the Romantic tradition is actively used by Mary Shelley only to illustrate its flaws more fully, albeit ironically. The provocative use of the antisentimental or Gothic mode as the *modus operandi* of this critique is as Thornburg suggests most effective in the use of the “apparent antagonist” who the reader sympathises with, but with whom they are “not free to sympathise openly” which is linked to the anxiety at the creature being in effect continually named “Monster”. Victor Frankenstein endures the most criticism as the Romantic element in the story exactly because the reader sees it reflected through the “Gothic, mirror image truths” that are contained within the creature's character, which continually pleads for sympathy. Therefore, even in the use of her characters, Mary Shelley is

continually placing Romanticism and the Gothic seemingly at odds but at some points they remain inextricably linked.

By setting up or incorporating aspects of Romantic idealism and the anxieties of the Gothic, often exactly at those points where they seem to overlap only to then destabilise or undermine them, Mary Shelley is able to transcend both genres. This is the point at which *Frankenstein* becomes “shockingly new” and it is at this point that the scientific elements within the novel become important. By trying to distance herself from both traditions of the Gothic and Romanticism, by trying to establish something different through questioning established ideals and as Botting states “elevating” both, she does in fact create, not only herself as an author, but also leads to concerns which retrospectively are inevitable concerns within many kinds of science fiction. It is also important at this point to attempt to trace precisely how her subtle navigation through two known territories brought her onto different ground. The scientific elements contained within *Frankenstein* in a sense blend in with Mary Shelley’s destabilising of Romanticism as Romanticism itself coincided with what could be termed the second scientific revolution.

This scientific revolution incorporates both the evolutionary and industrial aspects of the sciences. It seems therefore only natural that in her very thorough critique of Romanticism, Mary Shelley would necessarily extend that into scientific exploration as well. Aldiss argues that “the division between the arts and sciences had not then grown wide” (Aldiss, 30), and this notion of the thin separation between the arts and the sciences is also explained by Wordsworth’s ideas about the “Man of Science” standing side by side with the poet.<sup>8</sup> The close relationship between literary works and scientific discovery is shown particularly in the similar strategy that is employed by both scientists and Romantics to attract attention, as Golinski points out:

The [sublime] was frequently used at the time in discussions of philosophy and literary criticism. It named an effect deliberately cultivated by visual artists, poets and prose writers. [...] Scientific lecturers and writers sought to elicit feelings of the sublime in their audiences – just as visual artists and poets might – with various ends in view. (Golinski, 532)

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<sup>8</sup> Wordsworth, William. “Preface to Lyrical Ballads with Pastoral and Other Poems (1802)”. This is examined in the first chapter of this thesis.

Interestingly, even though scientists obviously had very different goals to those of literary artists, they used the same means. This method of utilising the sublime makes the science of this era into another Romantic enterprise, as for scientists the sublime was a way “to inspire further efforts to conquer nature, to subdue it to human knowledge” (Golinski, 532). This view of the scientific use of the sublime is particularly important in relation to Victor Frankenstein, as his entire motivation for creating hinges on his efforts to conquer the natural order, which is highlighted by his aim to “penetrate into the recesses of nature, and shew how she works in her hiding places” (M. Shelley, 30). Mary Shelley consequently treats the scientific presumption of having the right to penetrate or conquer nature in the same way as she treats Victor’s arrogant belief that he can become a deified creator, as the two aspects are yoked together into the same act. Throughout *Frankenstein*, in fact there is a sense in which Mary Shelley portrays the Promethean attempt to control nature as dangerous:

In her novel, thunderstorms and lightning appear as forces of destiny, partially identified with the monster created by Frankenstein, but also manifesting the relentless powers of nature itself. [...] Storms accompany each subsequent appearance of the creature as it demands restitution for its solitary state. (Golinski, 536)

Therefore, when one considers this explanation, it is not merely for dramatic effect that Frankenstein creates his creature “on a dreary night of November” (M. Shelley, 38) or that a lot of the plot occurs in conjunction with witnessing “a most violent and terrible thunder-storm” (M. Shelley, 24). The raging of nature accompanying the creature, or in other words the natural wrath or judgement brought to bear on Victor by this unification of his creation with uncontrollable nature, is because he dared to interfere in the natural order of life. This unification of nature and the creature is a reasonable argument as Mary Shelley consistently uses the creature to articulate the most severe judgement of Victor’s failure. As electricity is the tool by which Victor tries to gain his god-like status, it is in a sense poetic justice that nature seems to retaliate with thunderstorms and lightning. The incredible amount of natural electricity churned out by nature consistently within this plot also makes clear that Victor has also failed to subdue or conquer nature as he intended. Therefore, Victor fails not only in his Romantic ambitions, but also as a scientist as it is clear that nature retains a grasp on him; as discussed previously he dies from natural causes, thereby realigning the natural order of life ending inevitably in death. Arguably, it is yet again his egotistical attitude that Mary Shelley is deriding, combining Romantic egoism with scientific presumption showing how the one permeates the other in order to illustrate extensively how dire the consequences of such egotism can be.

Mary Shelley's unease with "penetrative" science, places *Frankenstein* in contrast with the Gothic, as it is concerned with a modern interrogation of the consequences of scientific overreaching. In addition, it is precisely for this reason that Mary Shelley's generic juggling is often claimed as the beginning of science fiction. Whether or not *Frankenstein* is in fact the first science fiction novel is a vast debate that falls outside of the concerns of this thesis, but it is interesting to take note of it because it shows how difficult it is to place *Frankenstein* securely in a specific genre. One of the reasons *Frankenstein* is appropriated by some to be the origin of science fiction, is that throughout her narrative Mary Shelley incorporates contemporary scientific debates, which she was aware of at the time. As Mellor explains:

The works of three of the most famous scientists of the late eighteenth and early nineteenth century – Humphry Davy, Erasmus Darwin, and Luigi Galvani – together with the teaching of two of their ardent disciples, Adam Walker and Percy Shelley, were crucial to Mary Shelley's understanding of science and the scientific enterprise. (Mellor 1989, 90)

The fact is that Mary Shelley did include these scientists' theories into her novel. Humphry Davy had a fascination with electricity that is mirrored by Percy Shelley's interest in it, and Davy in his public lectures and writings "listed electricity among the means by which man was enabled to 'interrogate nature with power'" (Golinski, 536). Erasmus Darwin's ideas on biological evolution, on the other hand, rest particularly on a more passive approach as he merely observed rather than interfered. Interestingly, Darwin is also invoked in the preface of the 1818 edition of *Frankenstein*:

The event on which this fiction is founded has been supposed by Dr Darwin, and some of the physiological writers of Germany, as not of impossible occurrence. (M. Shelley, 3)

Moreover, it is here that the major argument for Mary Shelley being the founder of science fiction emanates from. Aldiss' definition of science fiction adds credence to the theory:

Science fiction is the search for a definition of mankind and his status in the universe which will stand in our advanced but confused state of knowledge (science), and is characteristically cast in the Gothic or post-Gothic mode. (Aldiss, 25)

By arguing, that science fiction is cast in a Gothic or post-Gothic mode, Aldiss is in fact laying the foundation for an argument that can only conclude with *Frankenstein* as the origin of science fiction as his wording invokes precisely the type of manipulation that Mary Shelley uses with the Gothic and the Romantic genres within *Frankenstein*. Aldiss therefore sets the stage, and Freedman argues from that established point that the preface in itself states that Mary Shelley was writing science fiction, noting that by calling upon Darwin she is in fact celebrating his scientific pursuits, and he lays a lot of emphasis on the wording of the preface:

*Not of impossible occurrence:* these four words point to much of Mary Shelley's stunning originality and, in particular, to the way she decisively broke with the Gothic and other supernatural literary traditions by which she was so heavily influenced in order to invent science fiction. (Freedman, 255)

For Freedman, as well as for Aldiss, Mary Shelley breaks with the Gothic to invent a new genre, and it is the scientific elements she incorporates that leads them to this conclusion. Regardless of which way one might decide in regards to this debate, what is more important to contemplate is that the debate exists at all. What is apparent is that Mary Shelley is retrospectively seen as important for science fiction, as Botting emphasises that:

*Frankenstein* casts a gigantic shadow over science fiction: incessantly alluded to, repeatedly cited, regularly adapted and reworked, its presence is everywhere. (Botting 2008, 135).

For the purpose of this thesis, what is most significant is that this "gigantic shadow" or even the discussion of whether or not she invented a new genre highlights precisely how nuanced, subtle and original Mary Shelley's navigation of both Romanticism and the Gothic in fact is. The ripples, effects and debates caused by her loosing her "hideous progeny" upon the world have not yet ceased and have surpassed anything she would have expected.

Finally, it is of some importance to illustrate how this interplay between all of the discussed genres are important, for it would be an oversimplification to assume simply that the Romantic or Gothic elements are completely discarded in favour of the scientific elements that according to Freedman "breathe(s) rational, scientific atmosphere" (Freedman, 255) into *Frankenstein*. Interestingly all of these elements and genres seem able to coexist in *Frankenstein* and consequently make it, as Botting describes it a "hybrid fictional species":

The novel, a new and hybrid fictional species, begins in a ghost story competition but takes its bearings from the contemporary scientific endeavours [...] It looks back and lurches forward, like its monstrous protagonist, moving between the work of alchemists seeking the magical *elixir vitae* and the empiricism of new scientific ideas and techniques for understanding and transforming the physical world. Generically too, the novel is difficult to categorise: emerging from a context of Romantic companions, aesthetics and experiments, it abandons the supernatural events and superstitions of gothic fiction. (Botting 2008, 134)

It is precisely this "lurching" quality which makes it "difficult to categorise"; the fragmented piecing together of three genres into a cohesive and original text, which makes Mary Shelley's *Frankenstein* important in different ways to each genre that she assimilates. It is important in relation to Romanticism, in that it lends itself to an insightful contemporary critique of



Romanticism's flaws. It is important to the Gothic exactly because as Botting illustrates, it steps away from the Gothic, highlighting certain aspects of the Gothic more effectively in that created distance:

Gothic fiction, for all its wandering in desolate landscapes and invocation of diabolical forces, never strays far from home: it plays upon human fears and anxieties, its hauntings emanating as much from within as without. In their overlap, however, a long and interwoven association in the realms of modern popular literature and culture, both genres give form to a sense of otherness, a strangeness that is difficult to locate: monstrosity appears in the future and the past, in the mind and in culture at large, taking form in individual, social and textual bodies. (Botting 2008, 131)

Botting is arguing that in the overlap of rational, scientific atmosphere and the Gothic, where for instance Victor's rationality as a scientist coexists with those aspects of his character that make him a Gothic "titanic hero-villain", there is a certain enhancement at work. The implication is that this combination of elements draws attention to the "sense of otherness" or "monstrosity" already prevalent in Gothic fiction. Just as by using Romance and Gothic in the same text Mary Shelley elevates each with the interplay of the other, here Botting implies a very similar elevation, pointing towards an intensification of the Gothic elements of *Frankenstein* through the incorporation of scientific components, making the anxiety of transgression immediate and in a sense eerily resonant.

Returning then to the main premise, it is exactly this strange fluctuation within *Frankenstein* of specifically Romanticism and the Gothic that remains poignant and clear. For Mary Shelley, her criticism of Romanticism was personal as well as ideological, but her "blistering critique" is expressive of so much beyond Romanticism:

Romance, in its modern, eighteenth- century form at least, provided the matrix in which gothic and science fiction gestated. Monstrous in form, romance bred monstrosities. (Botting 2008, 132)

Ultimately therefore, Mary Shelley's first novel shows not only how masterfully she manages to negotiate various literary forms, but also, that at the centre of her story about a monster who is in actual fact not a monster but merely a creature, rejected and alone, the real monstrosity, the hideous monster that emerges from this story is in actual fact not only Victor Frankenstein and his Promethean ideals, but the idealist Romanticism that lies looming larger and more menacing behind those ideals. The monstrosity of Romantic idealism is that it alienates completely from society, lauding him as being destined for greatness that transcends the mundanely human. With striking clarity, Mary Shelley highlights that the horror of this lies in that it leads to an inability to

think or consider anything outside of the self, which presumes to heights that are better left unattained. Through Victor Frankenstein, it becomes increasingly apparent that such solipsism in fact leads to the individual regressing to something that is inhumane and monstrous rather than the transcendent genius this isolation is supposed to attain. The reality, which Mary Shelley continually brings to the fore, is that the Promethean sensibility or yearning, even when expressed through a scientist, leads only to tragic demise and breaches boundaries that are forbidden, and better left untouched. Romanticism, when seen in the harsh reality that Mary Shelley invokes, fails to bring any good to the world and more disconcertingly rejects redemption by being completely incapable of acknowledging responsibility for what it has in fact brought forth into that world. It is as Botting suggests the monstrous form breeding and birthing monstrosities. In creating her “hideous progeny” and entreating it to prosper as it will, Mary Shelley is highlighting that this progeny, like the creature, is only hideous because of what made it so.

## Conclusion

Alone, alone, all, all alone,  
Alone on a wide wide sea!  
And never a saint took pity on  
My soul in agony. ~ Coleridge, *The Rime of the Ancient Mariner*, (IV, 232-5)

Mary Shelley's dialogue with Romanticism and its ideology lasted longer than her relationship with Percy Shelley, who drowned in 1822. If one looks at her later work, specifically *The Last Man*, published in 1826, eight years after *Frankenstein*, it becomes clear that Mary Shelley was still criticising and questioning the principles of the Romantics even as she was mourning the passing of both Percy and Byron, who died in 1824. The concept of the title of *The Last Man* was something that Mary Shelley identified with, as her journal entry in May of 1824 reveals, "Yes, I may well describe that solitary being's feelings, feeling myself as the last relic of a beloved race, my companions extinct before me" (in Paley, vii). The isolation that Mary Shelley expressed so eloquently in the creature's narrative in her first novel has remained and intensified by virtue of becoming an actuality.

*The Last Man* therefore shows how yet again Mary Shelley dealt with the negative implications of Romantic idealism; consistently destabilising the ideal with a stark cynicism. The thematic similarities are evident, as Lokke argues that *The Last Man* "constitutes a profound and prophetic challenge to Western humanism" (Lokke, 116) by refusing to place humanity or its ideals as undeniably permanent or lasting. The fact that in *The Last Man* all except one man, the narrator, are systematically destroyed and that every one of the characters represents a certain aspect of Romantic ideology means that, as Lokke suggests, "*The Last Man* renders a devastatingly modern critique of the political, scientific, spiritual, and artistic aspirations of the post-revolutionary, Romantic era" (Lokke, 117). The distressing rejection of the creature in *Frankenstein* is thematically enlarged to the devastating isolation in *The Last Man* of Lionel Verney who is left all alone as the sole survivor of the human race. The creature is "borne away by the waves, and lost in darkness and distance" (M. Shelley, 191) and Verney, at the end of *The Last Man*, is seen adrift on the waves "around the shores of deserted earth" in a "tiny bark" (M. Shelley, *The Last Man*, 470). Their isolation is different circumstantially, yet the fact that they are both cast upon the ocean, endlessly drifting draws their fates closer together, each expressing in a certain sense a pervading loneliness. The difference between the self-imposed banishment of the creature and

the loneliness of Lionel Verney is that Verney's story is dedicated only to "the illustrious dead" (M. Shelley, *The Last Man*, 466), whereas *Frankenstein* retains the assumption that there are those who will benefit from hearing the cautionary tale of presumption unchecked. As Mellor points out this complete destruction of the human race except for the one who tells the story, points towards the fact that in *The Last Man*:

A Romantic ideology that grounds cultural meaning in the creations of the imagination or a dialectically developing phenomenological consciousness is destroyed by the disappearance of the human mind itself. *The Last Man* thus opens the way to twentieth-century existentialism and nihilism. (Mellor 1989, 169)

The transient reality of human consciousness and specifically the creations of the imagination are dealt with much more harshly in *The Last Man* as Mary Shelley destroys them in their entirety. The desolate ending of this *roman-à-clef* is all the more powerful because she invokes particularly those that she is mourning, and methodically tabulates their deaths. As Lokke implies:

Because the objects of Shelley's mourning are among the central literary and intellectual figures of her age – figures in conversation with an earlier generation of writers and thinkers – her *roman-à-clef* inevitably entails representations, both elegiac and profoundly bitter, of Percy Bysshe Shelley's idealism, Byron's titanism, Wordsworth's naturalism, Coleridge's aestheticism, and the progressive commitments of her parents, Mary Wollstonecraft and William Godwin. (Lokke, 117)

Where *Frankenstein's* cautionary element implies that there is still a choice to be made, a redemption that is possible if the warning is heeded – *The Last Man* intensifies the tragic hopelessness of aspiring to greatness by showing the fleeting nature of such idealism.

If anything, *The Last Man* shows that Mary Shelley did not in fact soften her views on Romantic principles and their inherent danger as she grew older. Romanticism continues to fail when it comes to the implication of its principles into her concept of reality. The tragic demise that is brought about by following Romantic ideology, in the mind of Mary Shelley, is magnified to being in part responsible for the destruction of almost all of humanity. This illustrates that Romanticism retains its monstrosity in the eyes of Mary Shelley. The Romanticism that Mary Shelley was familiar with isolated rather than integrated the individual. In her use of Romanticism, utilising and invoking it on the one hand only to undermine it or question its main tenets on the other, Mary Shelley illustrates the difficulties that are inherent in almost every aspect of an ideology that glorifies the individual and reveres the creative enterprise springing from such an idealised individuation above the communal or societal good.

In her introduction to the 1831 edition of *Frankenstein* Mary Shelley writes that “what terrifies me will terrify others” and in this confident assertion lies a key element of how Mary Shelley defined herself and explained her own insight or vision for her story. The implication of this is that it illustrates how Mary Shelley saw herself as part of or similar to others, a group or a collection of people. With this phrase, as seemingly insignificant as it might seem, Mary Shelley is confirming the humanity that she shares with “others”. There is no presumption of standing apart or more precisely above the human. Her expression of connection is opposed to the presumption of the creative Romantic, whose sole purpose is to glorify that which sets him apart. The presumption of genius, deification and being famously misunderstood by the masses which only adds to the lustre of standing outside of reality, is completely absent in this expression of inspiration. Therefore Mary Shelley’s affirmation of her connection to those around her reaffirms her ability to create. Her imagination springs from her understanding of humanity rather than being isolated from it. She therefore actively sets herself apart from the Romantic expectation, specifically in the way in which the creature, seeking companionship, articulates the importance of connection rather than isolation. The monstrosity of Victor Frankenstein hinges on the fact that he is unwilling to be a companion or to create a partner for his creation in his own inability to take responsibility for his creative act. Linking this to Percy Shelley only reinforces the suspicion that for Mary Shelley to be involved with someone as egocentric as Percy must have been devastatingly lonely. Moreover, it is this loneliness that resonates through both *Frankenstein* and *The Last Man* alongside the criticism of Romanticism as if to suggest that they are in fact inextricably linked. By isolating instead of incorporating individuals Romanticism sees community and relationships as being debilitating and limiting. *Frankenstein* proves that it is precisely Mary Shelley’s understanding of others and the relationships that exist between people that enriched her own imagination and arguably has led to the lasting validity of *Frankenstein*. In an ironic twist of fate Mary Shelley and her work has thus gained immortality and popularity arguably unmatched by any of the Romantics who formed the basis of her criticism.

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University of Cape Town





# **Mathematical and Computer Modelling of Heap Leaching at the Agglomerate Scale with Application to Chloride Leaching of Chalcopyrite**

by

**Alexey Cherkaev**

Thesis presented for the degree of  
Master of Science in Engineering  
in the Department of Chemical Engineering  
University of Cape Town

October 2010

## Declaration

I know the meaning of plagiarism and declare that all the work in the document, save for that which is properly acknowledged, is my own.

Signed.....  
Alexey Cherkaev

Date. 01.12.2010

University of Cape Town

## Abstract

Heap leaching is a low-cost technology to extract metals (primarily copper) from low-grade ores that has recently become the focus of industry, due to economic pressure. While this technology is well established for processing minerals such as cuprite or chalcocite, it is still under development to process chalcopyrite. As a result, there is a significant amount of investigation work around heap leaching of chalcopyrite. In this context, it has been suggested that chloride assisted leaching of chalcopyrite can achieve comparable rates relative to the more established bioleach technology at lower temperatures.

Heap leaching involves a number of transport and reaction phenomena that interact with each other. Therefore, investigation of such interactions requires a tool that can combine analysis of these phenomena to show their effects on the overall process. Mathematical modelling is considered to be an effective tool to study such complex systems and has been successfully applied to heap leaching. Various heap leaching models have been developed since 1974, paying attention to different phenomena such as solution flow, species transport and reaction kinetics, and heat transport.

The current work investigates the effect of an apparent mass transfer constraint in heap environments, inhibiting the chloride assisted leaching of chalcopyrite. It has been hypothesised that the transport of one of the reagents or products limits the process. A modelling approach has been chosen to test the hypothesis on the basis of a limited set of existing data. This data has been obtained from the experiments (conducted at Murdoch University, M. Nicol, 2010, personal communication) where potential measurements have been taken near the surface of chalcopyrite accessible for leaching via a capillary to mimic diffusion in pores or cracks in ore particles.

First, a simple steady-state model has been developed to provide an extensive analysis of the data from the "pore-diffusion" experiment. This model was calibrated using one set of that data and validated against the rest of that data. This approach shows that it is transport of the reaction product, cuprous ions, that limits the overall process. As an additional result, the values of the leach reaction rate coefficients have been obtained.

This model has then been extended by incorporating additional effects (such as time dependence, advection and mineral distribution) to simulate leaching at the agglomerate scale. This extension has enabled the investigation of micro-effects (such as pore diffusion) on the

overall leaching rate. Moreover, the model has been formulated to be more generic, i.e. it is capable to simulate different types of leaching by adapting specific reaction kinetic models, ore mineralogy and etc. The mathematical formulation of the model is quite complex, hence a fairly sophisticated numerical scheme and programme platform were developed. Extensive extensive sensitivity have been conducted to prove the robustness of the new numerical algorithm. However, the application of this model to chloride leaching of chalcopyrite, based on the calibration using parameters established in the initial study, has shown extremely slow rates, which appeared to be much slower than those found in preliminary column studies. This discrepancy indicates the process requires further study at the micro-scale to validate key model parameters such as the chalcopyrite reaction rate. An appropriate experimental programme has been proposed, which should be pursued in a follow-up study.

## Acknowledgements

I would like to express my gratitude to my supervisor, Dr Jochen Petersen, for his guidance in the engineering field, teaching me to trust engineers and his effort to understand me, a mathematician, and for his patience correcting my grammar mistakes.

Many thanks to my family and my friends for their support and encouragement. Without them the present text would never be written.

I would also like to thank BHP Billiton Ltd. for the financial and administrative support. Especially, I would like to thank Dr David Dew and Ian Harvey.

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# Nomenclature

General Symbols		
$\rho$	Density	kg/m <sup>3</sup>
$\nu_{ij}$	Stoichiometric matrix	—
$\varphi$	Order of the mineral leach reaction	—
$\varphi_i(x)$	Basis function in FE	—
$v$	Fluid velocity	m/sec
$C$	Species concentration	mol/L or mol/m <sup>3</sup>
$D$	Diffusivity	m <sup>2</sup> /sec
$S$	Source term	mol/(m <sup>3</sup> · sec)
$T$	Temperature	K
$x(x, y, z, l)$	Coordinates in the space	m
$X$	Mineral conversion	—
$r$	Radius (of a sphere or a cylinder)	m
$r$	Rate of reaction	mol/(m <sup>3</sup> · sec)
$t$	Time	sec
$L_2(\Omega)$	Lebesgue space of functions integrable with their squares	—
$w$	Test function	—
$n$	Normal vector to a surface	—
$\mathcal{S}(\Omega)$	A set or a space of functions	—
$K$	Stiffness matrix (FE)	—
$M$	Mass matrix (FE)	—
$F$	Source-term vector (FE)	—
$\mathcal{N}$	Species flux	mol/(m <sup>2</sup> · sec)
$R$	Universal gas constant	8.314 JK/mol

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Literature Review Specific Symbols

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$\phi$	Media porosity	–
$\mu$	Liquid viscosity	Pa · sec
$\theta$	Fluid saturation	–
$\varepsilon$	Volumetric fraction of void space	–
$\nu$ ( $\nu$ )	Flux	mol/(m <sup>2</sup> · sec)
$\Omega$	Domain	–
$\partial\Omega$	Boundary of the domain $\Omega$	–
$\psi(x)$	Form function in FE	–
$K$	Unsaturated hydraulic conductivity	m <sup>3</sup> · sec/kg
$p$	Pressure	Pa
$g$	Gravity acceleration	9.8 m/sec <sup>2</sup>
$N$	Microbial population	cells/m <sup>3</sup>
$f$	Some non-linear function	–
$\mathbb{R}$	Set of real numbers	–
$k$	Reaction rate coefficient	–

---



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Model Specific Symbols

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$\alpha, \beta$	As subscripts: indices, that denote species and reaction respectively	–
$\alpha, \beta$	As coefficients: recasting coefficients for reaction rates	–
$E$	Potential	V or mV
$E_a$	Activation energy	kJ/mol
$F$	Faraday constant	$9.64853 \times 10^4$ C/mol
$B$	Coefficient, that defines concentration, equation (3.16)	mol/m <sup>3</sup>
$p$	Leaching reaction rate coefficient	mol/(m <sup>2</sup> · sec)
$k$	Reaction rate coefficient	mol/(m <sup>3</sup> · sec)
$q$	Index that denotes the number of agglomerate	–
$H$	Height of the agglomerate	m

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# Chapter 1

## Introduction

At the present time the mining industry faces many challenges due to rising demand for metals, such as copper, low raw material prices and poorer ores currently available. These challenges force engineers and scientists to find effective low-cost processes to extract metals from ore. One of the most cost-effective processes is heap leaching as it avoids ore milling (it only requires crushing) and simplifies waste treatment. However, the largest concern about the process is its low effectiveness: metal extraction rarely reaches 80% and proceeds quite slowly, often over the course of years (Watling, 2006). The improvement of the heap leaching operations is one the main objective for research in this area.

A specific example of the successful application of heap leaching is the extraction of copper from oxide ores. As for copper sulphide ores, heap bioleaching has been proved to be effective for chalcocite ores. However, chalcopyrite ores remain the biggest challenge for the industry. As chalcopyrite ores are usually poor (with a very low mineral grade), current technologies (smelting or pressure leaching) are cost and energy intensive for extracting copper from these ores, making heap leaching the only available alternative. However, heap leaching of chalcopyrite ores is still under development, which implies high risks for industry to implement them. Therefore, extensive research in this area continues (Watling, 2006).

Bioleaching is acknowledged as the most promising method to treat copper sulphide ores in hydrometallurgy, including chalcopyrite. However, there are cases where this method is not applicable, for example, in the absence of pyrite which is necessary to maintain leach environments at elevated temperatures. Chloride assisted leaching (Majima *et al.*, 1985)



is considered as an alternative to bioleaching in these cases. Initial laboratory studies (in stirred tank reactors) have demonstrated reasonably high reaction rates (relative to heap leaching) with a high activation energy suggesting substantial acceleration at elevated temperatures (Velasquez-Yevenes *et al.*, 2010a). However, column tests that were conducted to demonstrate the application of the process have shown much slower rates, which did not increase significantly with the increase of temperature (Petersen, 2008).

## Hypothesis and objectives

This observation led to the postulation that the *leach process in a heap environment is governed by mass transfer limitations* — possibly the diffusion of reagent species (oxygen or the cupric/cuprous ion couple) through the porous micro-structure of the heap bed — rather than the reaction kinetics. This postulate has been supported by the preliminary analysis of column tests data (Petersen, 2008). A possible explanation of this effect has been proposed but has not been examined yet.

Furthermore, limited data on reaction kinetics existed at the time the present study commenced, and all of it has been derived from stirred tank work on chalcopyrite concentrate. In order to test the feasibility of above postulation before embarking on a lengthy laboratory study, it has been proposed to develop a micro-scale mathematical model of the diffusion-reaction process through a hypothetical ‘pore’ within the heap bed and test it using the available kinetic and thermodynamic data. The results of this model can help to identify gaps in current understanding of the leaching mechanism more precisely and, consequently, to guide further experimental work.

Additionally, it has been proposed to integrate a micro-scale model to macro-scale one to investigate the effect of micro-phenomena on the overall leach process. The ultimate goal of the macro-scale model is the analysis of data from column experiments.

Overall, objectives of this study can be summarised as follows:

- To determine whether diffusion effects indeed are rate limiting in the given context and whether it is the diffusion of oxygen or copper that limits the process;
- To find the implication of these phenomena on the overall rate of heap leaching using this process (according to the model results and within model assumptions);

- To identify the best way of conducting of further experimental work to further characterise and qualify the phenomenon.

## Thesis outline

The thesis consists of four main chapters:

**Literature Review** gives an overview of heap leaching technology and heap leach modelling. Particular attention is paid to the current state of understanding of the chloride leaching of chalcopyrite. Additionally, it gives an overview of numerical methods that are important in the context of heap leach modelling.

**Chloride Leaching Model** gives the analysis of the data from a simple pore diffusion experiment. A steady-state diffusion (micro-scale) model is developed to achieve this. At the end of this chapter, reaction rate coefficients are determined by calibration and validation of the model.

**General Modelling Platform Development** extends the model, developed in the previous chapter, bringing some generic properties into it (macro-scale model). As the model becomes more complex, more elaborate numerical techniques have to be employed which are discussed in detail. Model sensitivity with respect to various parameters is tested to determine whether the model provides robustness and effectiveness of the simulation.

**Conclusions and Further Research** outlines main results of the current study and formulates proposals for further experimental research.

## Scope and limitation

The present work is a modelling study, i.e. no own experiments have been conducted. Instead, this work has used existing experimental data and made suggestions for experimental research to be done in a follow-up study.

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# Chapter 2

## Literature review

### 2.1 Principles of hydrometallurgy

Hydrometallurgical processes revolve around the dissolution and removal of metallic compounds in aqueous phase. In comparison with conventional pyrometallurgy, it is a recent development, and only started in the late 19th century but enjoys increasing acceptance in industry. Advantages of hydrometallurgy include (Gupta and Mukherjee, 1990):

- The capital cost for smaller scales of operation of hydrometallurgical processes is lower than that of pyrometallurgical ones, which are feasible only at a large scale.
- Hydrometallurgical processes are more flexible in treating complex ores due to availability of a multitude of process routes for compound separation.
- They are perceived to be less energy consuming for low-grade ores.
- They have been suggested as an alternative processes to reduce air pollution commonly associated with pyrometallurgical processes.

However, hydrometallurgy has some disadvantages as well (Gupta and Mukherjee, 1990):

- Hydrometallurgical operations require higher levels of control, similar to chemical plants (for pressure leaching).
- Hydrometallurgy is less economically competitive for high-grade ores or concentrates.

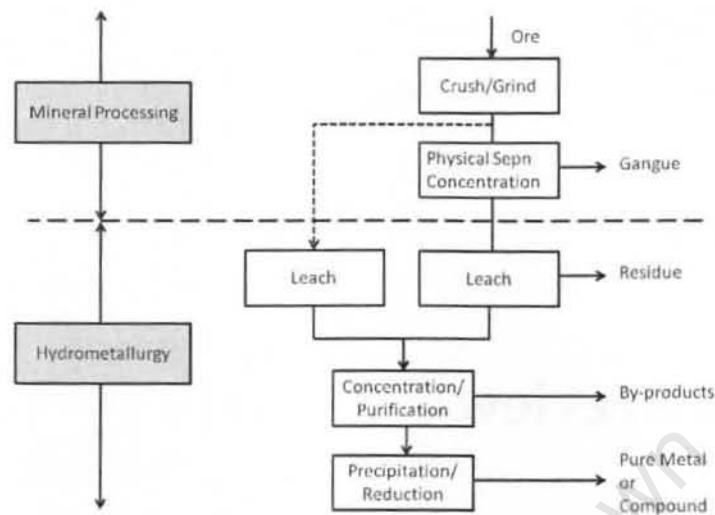


Figure 2.1: A generic hydrometallurgical process (adapted from Nicol (2007))

- Liquid and solid waste are still generated in the hydrometallurgical process. This waste may require special treatment for disposal.

A short overview of a typical hydrometallurgical process is given in Figure 2.1. Upstream processes, that consist of ore preparation for leaching, are known as mineral processing. Downstream processes include solution concentration and purification, followed by salt precipitation or metal reduction, resulting in a metallic compound or a pure metal, respectively.

### 2.1.1 Upstream: mineral processing background

Most processes applied to the ore, including hydrometallurgy, require pretreatment of the bulk material. First, ore needs to be extracted from the mine. The next step is milling followed by flotation aiming to produce a concentrate suitable for further treatment. If the mineral grade in the ore is low, milling becomes economically inefficient at some point, as it involves substantial consumption of energy in order to mill a large amount of bulk material, resulting in little concentrate produced. Alternative to milling is to crush the ore only

coarsely for heap leaching or it can be left completely uncrushed for dump leaching (Nicol, 2007).

### 2.1.2 Types of leach processes

The purpose of leaching hydrometallurgy is the dissolution of the desirable metal into solution. This can be done by using different reagents, depending on the mineralogy of the ore: acids, alkalies, soluble salts. A number of reactor technologies have been developed in order to get economical benefits depending on the grade and mineralogy of the ore. Such reactor technologies include (Gupta and Mukherjee, 1990):

***In situ leaching.*** One of the oldest types of leaching (it is reported to be used in 15th century in Hungary). The ore in this process is left unmined and leaching occurs within the ore body. The process has been found beneficial for extremely low-grade ores, where mining would be too expensive. The main advantage of this process is low capital cost, while difficult-to-predict output and potential contamination of ground water are listed as short-comings.

**Dump leaching** is commonly used for the low grade ores and run-of-mine (RoM) materials for which concentration appears not cost effective. Ore is mined (but it is neither milled nor crushed) and piled in dumps. Leaching solution is sprayed over the pile and allowed to drain through it. It is collected at the bottom through drainage systems and pumped to the refinery plant for further treatment. Often waste or run-of-mine ore is used for dump leaching. This low-grade ore had been dumped since it was perceived that treatment of such ore is non-profitable. However, as economical situation changes and technology evolves, dump leaching can appear to be an effective way to extract metals from it. The process can require long periods before metal dissolution becomes profitable. As a result, this process is beneficial only at a very large scale.

**Heap leaching** is another old process, that has been used at least since the 18th century to recover copper (the recover of copper from low-grade ores is a typical example of this type of leaching). In comparison with dump leaching more efforts are made to facilitate leaching: before the heap construction, ore is coarsely crushed and piled on a

specially prepared area. In addition to the presence of dissolved reagents, the process can require oxygen supply. In this case air is blown from the bottom of the heap into the ore bed. The result of the preparation work (and additional cost associated with it) is a significantly faster leach rate.

**Vat leaching** is close to heap leaching, but the ore is placed in large basins (vats) and completely immersed in leach solution. Due to higher costs involved and a lack of advantages against heap leaching this process is not longer practised.

**Agitated leaching** is used for ore concentrates (after milling and flotation) as a direct alternative to pyrometallurgy. Commonly it involves leaching under higher pressure, allowing the use of temperatures above the boiling point of water at normal atmospheric pressure, leading to higher reaction rates. Another advantage of the process is faster mass transfer in comparison with heap leaching. However, the operation involves high energy consumption (for milling and agitation) and is associated with higher costs. As a result, it is usually applied to high-grade ores, after concentration.

To summarise, leach processes offer the set of the reactor technologies that can serve under different conditions, depending on the environmental conditions, grade and mineralogy of the ore, and other factors.

### 2.1.3 Downstream: solution purification and electro-winning

To complete the description of a typical hydrometallurgical process, it is important to mention downstream processes. They include solvent extraction processes and electro-winning (Nicol, 2007).

Schematically, downstream processes for copper heap leaching are shown in figure 2.2. At the first stage copper ions from the aqueous solution are extracted to and concentrated in an organic phase, this is followed by the extraction these ions to aqueous phase again, that can be treated by electro-winning. Each circuit consists of number of solvent extracting stages operated counter-currently.

Pregnant leach solution (PLS) from the heap comes to the first solvent extraction (SX) stage in the loading circuit, where it is mixed with immiscible organic phase (usually kerosene

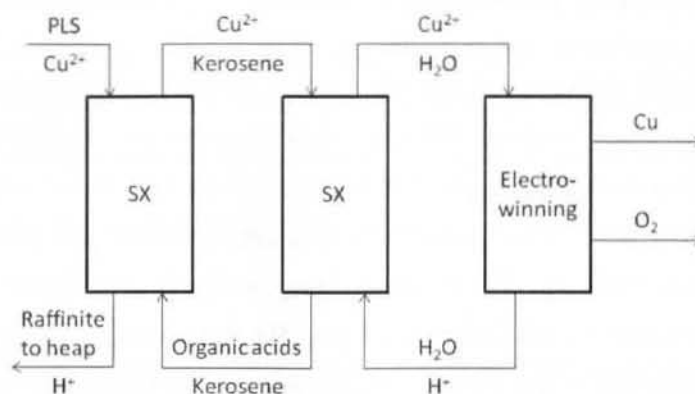
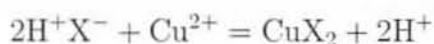


Figure 2.2: Solution purification and electro-winning for copper heap leaching

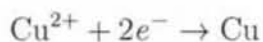
in which an organic complexing agent is dissolved) that cause copper ions from aqueous phase migrate into organic one by forming a stable compound with an agent dissolved there:



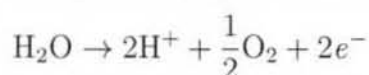
Protons  $\text{H}^+$  migrate in the opposite way: from the organic phase into the aqueous one. After that these phases are separated, aqueous phase is sent to the next stage of the loading circuit, while organic one goes into the stripping circuit. PLS proceeds through several SX-stages, decreasing copper concentration at each of them, after this it is sent back to the heap.

At the second (stripping) circuit, organic solution is mixed with acidic aqueous solution (again, in number of stages), resulting in copper ions migration into the latter. After separation, aqueous solution is sent to electro-winning and the organic phase comes to the bottom of the loading circuit (see figure 2.2).

At electro-winning plant copper is plated at the cathode:



While oxygen and protons are generated at the anode:



This results in the increase of the concentration of acid, which is required in the stripping circuit.



## 2.2 Heap leaching practice

The growth of the world demand for metals, such as copper, nickel and zinc compels the metals industry to process low-grade ores. However, conventional methods (such as pyrometallurgy and agitated leaching) can be economically inefficient for such ores due to the need to produce concentrates. This results in the application of low-cost processing methods: *in situ*, dump and heap leaching. For example, heap leaching of copper is used in various mines in Chile (Escondida, Cerro Colorado, Lince II) and Australia (Nifty Copper, Whim Creek and Mons Cupri) where the average grade of copper is from 0.3% to 1.8% (Watling, 2006).

Most of copper heap operations deal with oxide minerals (such as tenorite,  $\text{CuO}$ , or cuprite,  $\text{Cu}_2\text{O}$ ) using chemical leaching. As oxide minerals get depleted (typically 80% after 18 days), the operation turns to bioleaching to treat sulphide minerals, primary chalcocite,  $\text{Cu}_2\text{S}$ , and covellite,  $\text{CuS}$ , (which takes longer, about 200 days for 80% of chalcocite, Watling, 2006). In contrast to successful application of (bio-)leaching to chalcocite and covellite ores, leaching (and bioleaching) of chalcopyrite,  $\text{CuFeS}_2$ , still remains problematic, which forces researches to develop new technologies to treat this mineral (Watling, 2006).

## 2.3 Heap leaching of copper sulphide ores

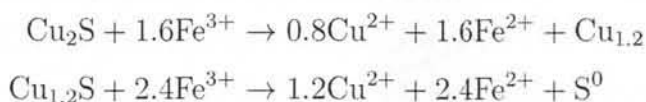
Heap bioleaching is the main technology to treat low-grade copper sulphide ores (with a mineral grade around 1%, so conventional methods are ineffective) that is used at the industrial scale. However, there can be cases where the application of bioleaching is not possible or associated with high costs. For example, if chalcopyrite ore does not contain pyrite,  $\text{FeS}_2$ , it is problematic to use bioleaching since the presence of pyrite is important to establish steady bacterial growth (Du Plessis *et al.*, 2007). These cases have led to the development of alternative technologies, such as chloride leaching.

### 2.3.1 Bioleaching

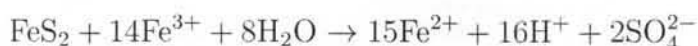
Sulphide minerals require oxidative reactions in order to leach metals (Watling, 2006). In comparison with non-oxidative leaching, these reactions proceed less rapidly. Additionally,

reagents can be consumed in larger quantities. In this case, a microbiological activity inside a heap can result in recovery of reagents leading to leaching facilitation.

For example, a typical reaction for chalcocite leaching, that proceeds in two steps (Petersen and Dixon, 2003), results in consumption of substantial amount of ferric ions:



Leaching of pyrite, that is often found with chalcocite, consumes ferric ions as well:



Micro-organisms can facilitate oxidative reactions that regenerate ferric ions from ferrous:



Additionally, they oxidise elemental sulphur, generating acid that can be consumed by gangue oxide minerals:



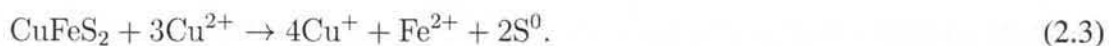
Since oxidative reactions (2.1) and (2.2) are exothermic ones, they result in a substantial increase of the temperature leading to higher reaction rates.

Overall, microbiological activity facilitates leaching through recovery of reagents and the temperature increase inside the heap.

### 2.3.2 Chloride leaching of chalcopyrite

It has been reported that the oxidative leaching of chalcopyrite is more effective in chloride solution than in sulphate solutions (Lundstrom *et al.*, 2005). The effect is explained by the higher rates of electron transfer in chloride solutions. Additionally, it has been suggested that there is formation of passivating products on the surface of chalcopyrite in sulphate solutions, whereas the chloride leach solution is free of these products (Lundstrom *et al.*, 2005).

In chloride media cupric ions act as oxidants to dissolve chalcopyrite:



In Al-Harashseh *et al.* (2008) it has been suggested that chalcopyrite dissolution occurs in two stages. First, it is oxidised by ferric ions:



This reaction generates cupric ions, that, in turn, dissolve chalcopyrite as the second stage. However, the dissolution rate by this mechanism has been reported as relatively slow. Since the addition of  $\text{CuCl}_2$  into solution media enhances the overall dissolution, the effect of reaction (2.4), in the presence of cupric ions, is insignificant.

Velasquez-Yevenes *et al.* (2010b) have reported that the rate of the chalcopyrite dissolution in the chloride environment strongly depends on the potential value (with increased rates with the potential value between 550 and 620 mV). At the same time, the rate is fairly independent from other parameters, such as pulp density, iron and copper concentrations, acidity and the chloride ion concentrations (Velasquez-Yevenes *et al.*, 2010a). Additionally, it has been reported, that rates under electrochemical potential control were much lower than those where potential had been controlled by the injection of air (or oxygen). The exact explanation of this fact still remains unclear. Stirred tank experiments, conducted at different temperatures, have shown significant increase in the leaching rate at elevated temperatures. The value of activation energy of the leaching reaction, 72 kJ/mol, has been determined from this experiments (Velasquez-Yevenes *et al.*, 2010a).

In addition to the research in reaction kinetics, some (limited) studies have been conducted to investigate mass transfer constraints that can become limiting factors in heap environments using data from column experiments. It has been proposed that mass transfer (specifically diffusion) limits the increase of the leaching rate in column tests at higher temperatures (Petersen, 2008): the analysis has shown that the activation energy of the leaching process in these tests was 31.2 kJ/mol instead of expected 72 kJ/mol (see figure 2.3). This analysis also suggests that it is the rate of copper leaching rather than mineral conversion that is consistent between different ore types. This further support the notion that the rate of chalcopyrite leaching is controlled by reactant or product transport rather than mineral reaction kinetics.

The discrepancy between stirred tank reactor and column test data is therefore a strong motivation for further study on mass transfer constraints in the heap system, as is described in the next section.

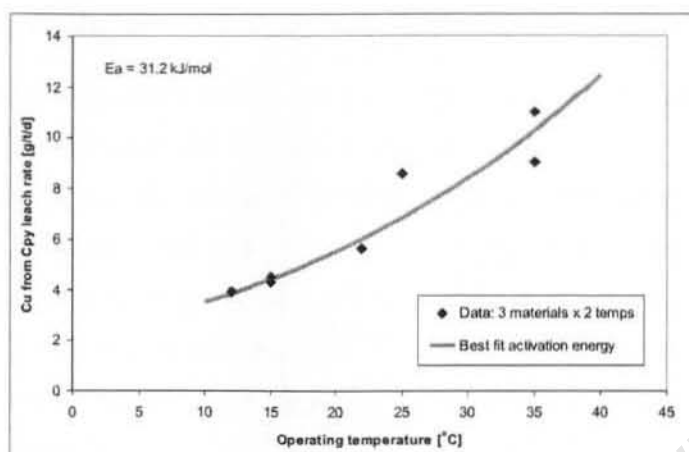


Figure 2.3: Activation energy of the leaching (adapted from Petersen, 2008)

### Capillary diffusion experiment

It has been acknowledged that chloride leaching of chalcopyrite proceeds at specific values of the potential: between around 550 and 620 mV (Velasquez-Yevenes *et al.*, 2010b). Outside of this potential window passivation has been observed (it has been reported that at low potential value chalcopyrite remained unleached and small quantities of covellite have been formed, the passivation at higher potential values has not been explained). For the chloride leaching the couple  $\text{Cu}^{2+}/\text{Cu}^{+}$  is believed to control the potential as represented by the Nernst equation:

$$E = E^0 - \frac{RT}{F} \ln \left( \frac{[\text{Cu}^{+}]}{[\text{Cu}^{2+}]} \right) \quad (2.5)$$

where  $E^0$  is the standard redox potential ( $E^0 = 162$  mV for the  $\text{Cu}^{2+}/\text{Cu}^{+}$  couple, Basson (2010)),  $R$  is the universal gas constant ( $R = 8.314$  JK/mol),  $T$  is temperature and  $F$  is the Faraday constant ( $F = 9.64853 \times 10^4$  C/mol) (Basson, 2010).

Mass transfer constraints can affect the potential, since they affect the ratio of  $[\text{Cu}^{+}]/[\text{Cu}^{2+}]$ . To investigate possible mass transfer constraints in the chloride leaching of chalcopyrite the following experiment has been conducted at Murdoch University, Australia (M. Nicol, personal communication, 2010, and similar work, Basson (2010), for covellite). The apparatus, shown on figure 2.4, has been constructed. It consists of a reservoir with the leach solution (0.2 M HCl and  $[\text{Cu}] = 0.5$  g/L), a capillary (diameter of 2 mm) of a fixed length (three

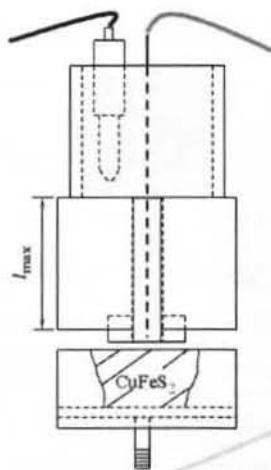


Figure 2.4: Apparatus for the pore-diffusion experiment (adapted from Basson (2010))

lengths have been investigated, 2 mm, 5 mm and 10 mm), which is connected to the reservoir, and a mineral, which is in contact with the leach solution at the other end of the capillary. Potential probes have been put into the bulk solution (reservoir) and in the capillary near the mineral surface, and the third probe is the mineral itself. Detailed description of this apparatus can be found in Basson (2010).

This experiment took about 40 hours to reach a steady-state. Potential values in the reservoir (reported remain unchanged) and near mineral surface at the steady-state are shown in 2.1. These results show a significant drop in the potential as the length of the capillary increases, suggesting the accumulation of cuprous ions near the mineral surface.

	Bulk	Pore Lengths		
		2 mm	5 mm	10 mm
Redox Potential	682 mV	647 mV	634 mV	622 mV

Table 2.1: Potential values in the reservoir and near mineral surface at steady-state

### 2.3.3 Physical principles of heap operation

Physically, heap leaching operation consists of several transport phenomena: solution flow, species advection and diffusion, air flow and heat transport.

As solution is injected into the heap, it starts moving downwards resulting in the advection of dissolved species, that can diffuse and disperse in the ore bed as well. In heap bioleaching air is pumped from the bottom of the heap so there are additional transport phenomena in a gaseous phase. If heat is generated by exothermic chemical reactions, it spreads through the heap and leaves the heap by means of conduction, convection, advection and radiation (Dixon, 2000).

A packed ore bed offers a porous matrix, i.e. a solid medium with a network of void pores. Solution applied to the surface of the heap migrates through these pores, spreading in the heap but keeping the main direction downwards. As the diameter of the individual pore is quite small, solution transport involves capillary effects, resulting in some portion of solution being stagnant, especially in dead pores that have limited connection with the main network (Bear, 1976).

Since pores usually are not fully saturated by the moisture, void space in pores forms a network for gas transport. If gas transport is not forced by the gas injection (higher pressure) at the bottom of the heap, it occurs via natural convection due to the difference of gas densities at temperatures inside the heap. Taking into account non-uniform permeability of the heap and the relative effects of forced and natural convections these effects lead to highly complex patterns of gas movement (Sidborn *et al.*, 2003).

Although reagents, either dissolved in the solution (such as metal ions) or in the gas phase (such as oxygen), are mainly transported in the respective bulk phase, they can diffuse, disperse and be adsorbed by the solid medium (diffuse into little cracks and pores inside ore particles) due to the fact that fluid transport is slow. Therefore, diffusion effects make a significant contribution.

Heat flow in heaps is one of the most complex phenomena since it can be transported in a number of ways. Firstly, heat can spread by conduction through rock and liquid. Secondly, it is transported with moving solution and gas (primarily in the form of water vapour). Depending on the zone in the heap, either water vapour condenses, or water evaporates resulting in the heat exchange between phases. Additionally, heat leaves the heap at boundaries: at



the bottom with moving solution, at other boundaries with moving gas phase and due to radiation (Dixon, 2000).

All physical phenomena described above are coupled with chemical phenomena, since chemical reactions leads to

- Consumption or production of reagents, thereby creating concentration gradients.
- Generation or consumption of heat creating local and regional temperature gradients, which impact on transport, especially of gas.

Overall, the interaction between physical, chemical and biological effects makes it impossible to study the impact of each effect on heap leaching in isolation from others. This results in a need to make a careful plan of experimental work in order to obtain meaningful and reliable data, and to use modelling as a tool since it can decrease the amount of experimental work and generate data that cannot be obtained directly from experiments. Thus, it forms a part of a cyclic process, where experimental work generates data that is then simulated by models. The results of model simulations are used to restate certain hypotheses, leading to redesigned experiments, followed by the generation of data from these experiments and so the cycle continues (Petersen, 1998).

## 2.4 Heap leach modelling

### 2.4.1 Mathematical base

Heap leaching modelling covers many aspects of the process. Different classes of models have been developed, emphasising different phenomena, such as hydrology problems, species transport, reaction models, etc. Each class narrows the focus on one effect, which is assumed to be overall rate limiting for the process.

Heap hydrology (bulk flow) models rely on flow through porous media which has been largely described by Bear (1976). This has been used in the modelling of groundwater flow (Freeze and Cherry, 1979) but recently has found application in heap leaching as well (for example Pantelis *et al.*, 2002; Bennett *et al.*, 2003; Cariaga *et al.*, 2005).

The dynamics of flow is described by the continuity equation, taking into account that solution does not occupy the entire space (Bear, 1976):

$$\frac{\partial(\phi\rho_\gamma\theta_\gamma)}{\partial t} + \text{div}(\rho_\gamma\mathbf{v}_\gamma) = 0 \quad (2.6)$$

where  $\phi$  is the media porosity,  $\rho_\gamma$  refers to the density of the fluid  $\gamma$  (for solution  $\gamma = w$ , for air  $\gamma = n$ ),  $\theta_\gamma$  is the saturation ( $\theta_w + \theta_n = 1$ ), and  $\mathbf{v}_\gamma$  denotes the velocity (specific discharge) of the fluid  $\gamma$ . Darcy's law is used to relate velocity and the fluid pressure:

$$\mathbf{v}_\gamma = -K \text{grad} p_\gamma \quad (2.7)$$

where  $K$  denotes the unsaturated hydraulic conductivity, which is a function of the saturation of the fluid. There are a number of semi-empirical models that relate fluid pressure to saturation (Bear, 1976). Any particular choice depends on the specific conditions being considered.

Since the solution inside particle pores remains stagnant, species transport is due to diffusion only. Therefore, the diffusion equation is used to describe the transport problem. A source term is usually involved in the equation, as mineral leaching reaction occurs inside particles.

Usually, particles are assumed to be spherical and spherically symmetric (Dixon and Hendrix, 1993b; Ogbonna, 2006). The diffusion equation for this system is as follows:

$$\frac{\partial C}{\partial t} = D \left( \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right) + S, \quad (2.8)$$

where  $D$  is the diffusivity and  $S$  refers to the source term.

Modelling at the agglomerate scale aims to combine effects of bulk flow and species diffusion, thus filling the gap between heap and particle scale phenomena. As a result, the interaction between bulk flow and particle diffusion is under consideration.

Agglomerates are formed by a number of ore particles, packed closely to each other, such that solution inside the agglomerate is virtually stagnant. Thus, a typical agglomerate system consists of two or three phases: flowing solution (and gas), stagnant solution (omitted in some models) and ore particles. Mathematically, agglomerate systems are described by some combination of equations (2.6) and (2.8) which varies depending on to what extent the agglomerate scale is considered in the overall heap model. This is elaborated in the next section.



### 2.4.2 Model approaches

There are two main groups of heap leach models: abiotic, that do not consider microbiological activity and associated effects, and bioleach models. The former group deals with either cyanide or oxide leaching, whereas a main focus of the latter lies in leaching of sulphide minerals.

#### Abiotic models

The New Mexico Cu oxide model (Roman *et al.*, 1974) has been reported to be the first approach to heap leach modelling (Dixon, 2003). This model aims to predict metal recovery based on the mean size of the ore particles. It uses a combination of product-layer diffusion controlled shrinking core kinetics for ore particles and plug-flow solution transport to estimate acid consumption for copper leaching from oxide ores. The model has been validated against experimental data using two column tests: the model, calibrated with data from the first column, has been validated against the data from the second column, that has been operated with similar parameters except the column height and width (Roman *et al.*, 1974). Another two models (U. Utah Cu oxide and SMP thin-layer leach models, as reported by Dixon (2003)) use similar shrinking-core formulation, but add some extra effects, such as "surface deposits", mineral grains that are accessible at the particle surface, and an additional stagnant solution phase.

The UNR (University of Nevada, Reno) general model (and similar UAMI gold cyanide model, Dixon, 2003; Dixon and Hendrix, 1993a) comprehensively considers diffusion and chemical effects. Ore particles are modelled as porous spheres with mineral deposits accessible either at the particle surface or inside particles via diffusion (equation (2.8)), solution plug-flow is assumed. Mineral dissolution is expressed by:

$$\frac{dC_{\text{mineral}}}{dt} = -kC_{\text{mineral}}^{\varphi}C_A \quad (2.9)$$

where  $\varphi$  is the reaction order and  $C_A$  is the reagent (acid) concentration. Thus, the reaction rate depends on the concentration of the single reagent and the availability of the mineral to leaching. It should be noted, that the model can take into account the presence of several minerals. Therefore, it enables the investigation of competitive leaching. The main model

limitation is the use of a single reagent. The model has been validated against experimental data and has shown consistent agreement with experimental data.

Cariaga *et al.* (2005) have developed a comprehensive model for acid leaching of copper. The main focus of this model is on the solution and species transport through the heap, whereas reaction kinetics is expressed using first order reaction kinetics. It uses the mass balance equation (2.6) for solution and gas phases. In order to relate fluids pressure to their saturations, this model uses the Brooks-Corey expression:

$$p_n - p_w = p_d \left( \frac{\theta_w - \theta_{wr}}{1 - \theta_{wr}} \right)^{-1/\lambda_{BC}} \quad (2.10)$$

where  $\theta_{wr}$  is residual water saturation and  $p_d$  is the atmospheric pressure. However, it is unclear from the model description what form has been used for hydraulic conductivity: either it has been expressed as a function of saturation (as a usual approach) or it has been assumed constant.

The information about the moisture content and the velocity distribution is used for the species transport equation:

$$\frac{\partial(\phi\theta_\gamma C_\alpha^\gamma)}{\partial t} + \text{div}(C_\alpha^\gamma \mathbf{v}_\gamma - \phi\theta_\gamma \mathbf{D} \nabla C_\alpha^\gamma) + \Phi_\alpha^\gamma = 0, \quad (2.11)$$

where  $C_\alpha^\gamma$  is the concentration of species  $\alpha$  in the phase  $\gamma$ ,  $\mathbf{D}$  is the dispersity-diffusion tensor, and  $\Phi_\alpha^\gamma$  denotes the source term. The model has not been verified against experimental data.

A similar model has been formulated in Sheikhzadeh *et al.* (2005), except that it considers only the liquid phase, whereas gaseous phase is assumed to be at atmospheric pressure. The correlation, similar to (2.10), has been used for pressure-saturation relation (van Genuchten's soil-water retention curves (SWRC)):

$$p_w = \frac{1}{\alpha} (\theta_e^{-1/m} - 1)^{1/n}$$

where  $\alpha$  and  $n$  are parameters of SWRC,  $m = 1 - 1/n$  and  $\theta_e$  is effective saturation defined as:

$$\theta_e = \frac{\theta_w - \theta_{wr}}{1 - \theta_{wr}}$$

Hydraulic conductivity  $K$  (see equation (2.7)) is expressed as a function of liquid saturation in the form:

$$K = \frac{k_i \rho_w g}{\mu_w} \theta_e^{1/2} \left( 1 - (1 - \theta_e^{1/m})^m \right)^2$$

where  $k_i$  is the bed intrinsic permeability that depends on the structure of the heap only,  $g$  is gravity acceleration and  $\mu_w$  is the liquid viscosity. The model has been tested against experimental data. It has been shown that model results follow general trends of the data, however, some effects, observed in experiments, have not been reflected in numerical results. Overall, model results has shown good correlation with experimental data with discrepancy not exceeding 10%.

Although recent abiotic models (Cariaga *et al.*, 2005; Sheikhzadeh *et al.*, 2005) involve a comprehensive description of solution (and air) transport, their application has limited capabilities. Firstly, they only consider uniform bed permeability and unifrom solution irrigation. As a result, model outcomes (saturation and concentration profiles) do not show significant variation in the horizontal direction (the only horizontal effect they incorporate is the effect of heap boundaries). Therefore, despite formulated as two-dimensional, these models are, effectively, one-dimensional. Secondly, the process description in these models is rather limited: they consider only oxide leaching using one reagent and one mineral.

### Heap bioleaching models

One of the first attempts of bioleaching models is the Kennecott Cu dump leach model (Dixon, 2003; Murr, 1980). The model is based on the combination of oxygen balance and shrinking core approach. As it was pointed out by Dixon (2003), this model considers pyrite leaching, instead of copper leaching, but it is assumed that pyrite and copper sulfide leaching rates are correlated.

Australian Nuclear Science and Technology Organisation (ANSTO) has developed a group of models since 1986 (Davis and Ritchie, 1986). Early models developed in this group considered oxygen transport as the only significant resistance for leaching. Similarly to the Kennecott model, these models rely on the correlation between copper recovery and oxidation of pyrite. There have not been any data presented to support the model (Dixon, 2003). The latest ANSTO model has been presented in Pantelis *et al.* (2002) as a set of equations that exploit a Darcian approach (2.7) for air and water transport. Again, this model considers only oxygen transport as a rate limiting factor. Microbiological activity has been considered in a very limited way, i.e. through oxygen consumption, but bacterial growth, attachment and other effects have not been included. As a result, this model in general is similar to abi-

otic models of Sheikhzadeh *et al.* (2005) and Cariaga *et al.* (2005). Additionally to previous models, this model incorporates heat transport. In simplified form, it looks as follows:

$$\bar{c} \frac{\partial T}{\partial t} + \overline{\rho c v} \cdot \nabla T = \nabla \cdot (\bar{k} \nabla T) + S_r - S_v \quad (2.12)$$

where  $\bar{c}$  is a combined conductivity of the ore bed (including gas, liquid and solid phases),  $\overline{\rho c v}$  denotes a combined advection term,  $\bar{k}$  is a combined coefficient of thermal conductivity,  $S_r$  is the heat generated by chemical reaction and  $S_v$  denotes the latent heat from water vapourisation (Pantelis *et al.*, 2002). Although the model is explicitly stated in the form of the set of equations, it remains unclear if it has been implemented (Dixon, 2003) since there is a lack of results presented.

A 2-D model has been presented by Leahy *et al.* (2005). However, the model description is somewhat vague and makes some unstated assumptions. For example, quite unusually for heap leach modelling, it uses Navier-Stokes equation to describe a gas flow:

$$\frac{\partial}{\partial t} (\varepsilon \rho v) + \varepsilon \rho \nabla \cdot (v v) = -\varepsilon \nabla p + \varepsilon \nu \nabla^2 v + \varepsilon B$$

where  $\varepsilon$  is a volumetric fraction of void space,  $\rho$  is gas density,  $v$  is gas velocity,  $p$  denotes hydrostatic pressure,  $\nu$  is viscosity and  $B$  is a body force that represents the resistivity of the porous media. It should be noted that the second term on left hand side of the equation (convective part of the time derivative) has an unusual form. Moreover, it is not clear what kind of product is meant there. The normal form of this term is

$$v \cdot \nabla (\varepsilon \rho v)$$

Although the importance of 2-D models and advances they have over 1-D ones have been pointed out, the subsequent development of this model (Leahy *et al.*, 2007) considers only 1-D problems. Moreover, it has abandoned the use of Navier-Stokes equation in favour of the plug-flow assumption.

Overall, in contrast to advanced development of abiotic models, bioleaching models still exploit oversimplified approaches resulting in neglecting of large amount of effects. One of the most comprehensive bioleaching model, HeapSim, is described in more details in the next section.

### HeapSim model

Being one of the major development in bioleach modelling, the HeapSim model is considered in somewhat more details in this section. The combination of diffusion, advection, heat transport, chemical and biological phenomena makes it the most sophisticated (and, at the same time, complicated) model.

The whole system is divided into (porous) solid phase, flowing solution, bulk solution that stays at dead pores and free space occupied by air. Dissolved species (both chemical and biological) are transported with the flowing phase via advection and in the stagnant solution via diffusion. Since the model can be adapted to different types of processes (i.e. for copper leaching from chalcocite or pyrite, or for zinc leaching, see Petersen and Dixon (2007b)), a general description is given below with some examples on copper bioleaching from chalcocite ore.

Solution is assumed to flow with the constant superficial velocity downwards. Species are transported with the solution flow in the vertical direction whereas diffusion moves them through bed pores in horizontal direction. The transport concept can be summarised in two equations:

$$\frac{\partial C_f}{\partial t} = -\frac{u_f}{\varepsilon_f} \frac{\partial C_f}{\partial z} + S_z \quad (2.13)$$

$$\frac{\partial C}{\partial t} = D \left( \frac{\partial^2 C}{\partial r^2} + \frac{\mu_s}{r} \frac{\partial C}{\partial r} \right) + \frac{S}{\varepsilon_s} \quad (2.14)$$

where  $C_f$  and  $C$  are concentration of chemical or biological species in flowing and bulk solutions respectively.  $u_f$  is the superficial velocity of the flowing solution,  $\varepsilon_f$  is the ratio of the flowing solution volume to the whole bed volume,  $S_z$  represents the flowing source term (described below),  $D$  is effective diffusivity,  $\mu_s$  is a shape factor: it equals to 0 for rectangular system, 1 for cylindrical and 2 for spherical.  $S$  is the source term for the bulk solution due to chemical reactions.  $\varepsilon_s$  is the ratio of the mass of bulk solution to that of solids (Petersen and Dixon, 2007a).

Equation (2.14) requires boundary conditions to complete the problem statement. They are derived for the cylindrical system as follows: solution flows around a cylinder of the radius  $R$  and can diffuse inside it. Thus, at the centre of the cylinder there is no diffusion:

$$\left. \frac{\partial C}{\partial r} \right|_{r=0} = 0$$

The outer boundary condition describes the exchange between bulk solution and flowing solution, i.e. it is incorporated into term  $S_z$ . The common practice to describe the exchange is to use the gradient of the concentration:

$$S_z = \kappa \frac{\partial C}{\partial r} \Big|_{r=R} \quad (2.15)$$

where  $\kappa$  incorporates some constants in order to match the dimension of equation (2.13).

In order to keep the mass balance of the system, that can be dislocated by the numerical scheme, there has been an additional effort to calculate the gradient in equation (2.15) with high accuracy: equation (2.14) is integrated over the space leading to the expression (detailed derivation can be found in Ogbonna *et al.* (2005))

$$\frac{\partial C}{\partial r} \Big|_{r=R} = \frac{1}{DR} \int_0^R \left( \frac{\partial C}{\partial t} - \frac{S}{\varepsilon_s} \right) r \, dr \quad (2.16)$$

It should be noted that a more precise evaluation of the integral is done at the expense of additional calculations required to evaluate the integral (2.16).

The source term  $S$  represents species consumption or generation due to chemical reaction or microbial oxidation, therefore, for any species  $i$  it has a general form

$$S_i = \sum_j \nu_{ij} r_j$$

where  $\nu_{ij}$  is a stoichiometric matrix of reactions and  $r_j$  is the rate of reaction  $j$ . Generally, all reactions can be split into two groups: those associated with mineral leaching and those connected with microbiological activity. Mineral leaching rate can be described by the equation (Petersen and Dixon, 2007a):

$$r_j = \kappa' \frac{\partial X_j}{\partial t} = \kappa' k_j(T) f_j(C) W_j(1 - X)$$

where  $\kappa'$  incorporates all necessary constants to match the dimension of equation (2.14),  $X_j$  is the conversion of mineral  $j$ ,  $f_j(C)$  expresses the dependence of the rate on the concentration of dissolved species that can facilitate or inhibit reaction, and function  $W_j(1 - X)$  describes the availability of the mineral surface for leaching.

The rate of microbial oxidation has a form

$$r_j = \frac{1}{y} \frac{dN}{dt} + k_m(N)$$



where  $y$  denotes the specific cell yield, i.e. the ratio of number of cells generated to mole of oxidation product.  $N$  is the microbial population,  $k_m(N)$  is the maintenance rate. The rate of change of microbial population is stated to be (Ogbonna *et al.*, 2005):

$$\frac{dN}{dt} = N \cdot \{\text{growth term} - \text{death term}\}$$

The growth term is expressed as a multiplication of monod-type expressions, each describing the effect of the particular factor on the microbial growth (more on growth term expressions can be found in Ojumu *et al.*, 2006). The death rate is stated to be dependent only on the temperature (Petersen and Dixon, 2007a).

Although the model in general is unsteady-state, gas transport phenomena are assumed to be under steady-state conditions (Ogbonna *et al.*, 2005). In the model the only species that can be present in the gaseous phase as well as liquid phase, is oxygen. Its transport in gaseous phase is described by the steady-state advection equation:

$$\frac{\partial p_{O_2}}{\partial z} = \kappa r_{O_2}$$

where  $p_{O_2}$  is the partial pressure of oxygen,  $\kappa$  is a proportionality coefficient and  $r_{O_2}$  is the rate of oxygen consumption in the system. From the gaseous phase oxygen can migrate into solution according to Henry's law:

$$C_{O_2}^* = k(T, C)p_{O_2}$$

where  $C_{O_2}^*$  is an equilibrium concentration. The rate of oxygen migration into solution is modelled as

$$r_{O_2} = k_L a (C_{O_2}^* - C_{O_2})$$

Since the model assumes steady-state conditions, this rate must be equal to the net oxygen consumption via reactions (2.1) and (2.2):

$$r_{O_2} = \text{net oxygen consumption}$$

The heat transport sub-model in HeapSim remains one of the most comprehensive for heap bioleaching. It accounts for heat conduction in solids and stagnant solution, advection

by flowing solution and gaseous phase, generation and consumption by evaporation and condensation, generation by chemical reactions and loss of the heat due to radiation at the top. Since heat transport involves many phenomena of different nature, its mathematical description is quite complex and is thus omitted here (details can be found in Dixon, 2000).

### 2.4.3 Model calibration and validation

There is quite a limited amount of information in the literature about calibration of heap leaching models. The most extensive explanation is given by Petersen and Dixon (2007b) for the zinc heap bioleaching model. This section shortly summarises that study.

In the model calibration study of Petersen and Dixon (2007b) model parameters have been divided into several groups: physical, fundamental, empirical. Physical parameters refer to those that are operator-selected and usually vary with application. For example, heap height, flow rates, ore characteristics, etc. Fundamental parameters are those material constants, that are available in the literature and usually do not depend on a particular application. They include the choice of mineral kinetic models, diffusivities, etc. Empirical parameters refer to rate constants and other parameters that can vary, depending on the ore mineralogy or structure. Although some of them can be found in the literature, most of them need to be determined from specially designed experiments. Since empirical parameters are least available, model calibration was focused on adjusting these parameters to fit experimental data.

Calibration was performed in several steps, improving the fit of model outputs to experimental data. However, the "fit" was considered on the intuitive level (i.e. how close points from a particular graph are to points from the experiment), instead of formalising it by introducing an error estimation. After four steps of parameter adjustments, model output and experimental data showed good intuitive fit.

Once calibration work was completed, the model was validated against data from a heap test, that has different physical parameters, including ore mineralogy. It was shown, that a significant difference in mineralogy can affect values of rate constants. Thus, the result of a direct comparison can have a limited significance. Despite this limitation, model has shown close fit with the data from the heap test.



## 2.5 Overview of numerical methods

Core equations that express the dynamics of the heap leaching can be reduced to those describing flow through porous media (2.6) and diffusion (2.8). Generally, these equations are non-linear, resulting in infeasibility to solve them analytically. Therefore, numerical techniques form a vital part of each modelling approach.

To illustrate how equations outlined in the previous section can be solved numerically, unsteady-state diffusion (or heat conduction) equation on the 3D domain is considered:

$$\frac{\partial C}{\partial t} - \frac{\partial}{\partial x} \left( D \frac{\partial C}{\partial x} \right) - \frac{\partial}{\partial y} \left( D \frac{\partial C}{\partial y} \right) - \frac{\partial}{\partial z} \left( D \frac{\partial C}{\partial z} \right) - f = 0. \quad (2.17)$$

This partial differential equation (PDE) requires initial and boundary conditions (BC):

$$\text{for } (x, y, z) \in \Omega \quad C(0, x, y, z) = C_0(x, y, z), \quad (2.18)$$

$$C \Big|_{(x,y,z) \in \partial\Omega_1} = C^*, \quad (2.19)$$

$$\left( -D \frac{\partial C}{\partial x}, -D \frac{\partial C}{\partial y}, -D \frac{\partial C}{\partial z} \right) \Big|_{(x,y,z) \in \partial\Omega_2} = (\nu_x, \nu_y, \nu_z), \quad (2.20)$$

Where  $\Omega$  is a domain,  $\partial\Omega = \partial\Omega_1 \sqcup \partial\Omega_2$  is its boundary,  $C^*$  is a specified value at one part of the boundary (Dirichlet BC), and  $\nu = (\nu_x, \nu_y, \nu_z)$  is a specified flux at another part of the boundary (Neumann BC).

Since this equation involves first-order time derivative and second-order space derivative, its solution can be performed in two ways: spatial derivatives can be discretised first, yielding an ordinary differential equation (ODE) with respect to time, or time can be discretised first, yielding a PDE with derivatives with respect only to spatial variables. The following sections show the solution of this equation, using the first approach, i.e.:

- Space discretisation;
- Time discretisation;
- Solution of non-linear system;
- Solution of linear system.

### 2.5.1 Finite differences

Finite differences (FD) is the simplest method to solve a PDE. The main advantage of this method is low computational cost for simple cases, such as rectangular domains and regular meshes (Bakhvalov *et al.*, 2006). FD has been used in the HeapSim model (Petersen and Dixon, 2005) and some other heap leaching models (Pantelis *et al.*, 2002; Ogbonna, 2006)

The FD method is illustrated on the simplified one-dimensional steady-state form of equation (2.17) on the interval  $(0, 1)$  as FD can be fully understood in this simplified case:

$$\frac{d}{dx} \left( D \frac{dC}{dx} \right) + f = 0 \quad (2.21)$$

with boundary conditions:

$$C \Big|_{x=0} = C^*, \quad (2.22)$$

$$-D \frac{dC}{dx} \Big|_{x=1} = \nu. \quad (2.23)$$

The term  $f$  is the source term, it can be a function of  $x$  or  $C(x)$  or both:  $f = f(C, x)$ .

In order to emphasise the principles of the method, the term  $D$  in equation (2.21) is assumed to be a constant. The segment  $[0, 1]$  is divided into  $N$  equal sub-segments  $[x_i, x_{i+1}]$ ,  $i = 0, \dots, N-1$  with length equals to  $\Delta x = 1/N$ . The second derivative is approximated by the expression:

$$\frac{d^2C}{dx^2} \Big|_{x=x_n} = \frac{C_{i-1} - 2C_i + C_{i+1}}{\Delta x^2} + O(\Delta x^2),$$

where  $C_i$  refers to the value of  $C(x)$  on the node  $x_i$ . Substitution of this expression into equation (2.21) leads to the set of algebraic equations

$$\frac{D}{\Delta x^2} (C_{i-1} - 2C_i + C_{i+1}) + f_i = 0, \quad (2.24)$$

for  $i = 1, \dots, N-1$ . This set of equations can be rewritten in a matrix form:

$$\mathbf{K}\mathbf{C} + \mathbf{F} = 0, \quad (2.25)$$

where  $K_{i,i-1} = K_{i,i+1} = D/\Delta x^2$ ,  $K_{i,i} = -2D/\Delta x^2$ , and  $K_{ij} = 0$  if  $|i - j| > 1$ .

Boundary condition (2.22) is naturally projected on the mesh:

$$C_0 = C^*$$

As for the boundary condition (2.23), there are two ways of dealing with it. The first way is to use  $x_{N-2}$ ,  $x_{N-1}$  and  $x_N$  to discretise the derivative, keeping the second order of discretisation:

$$\left. \frac{dC}{dx} \right|_{x=1} = \frac{-\frac{1}{2}C_N + \frac{2}{3}C_{N-1} - \frac{1}{6}C_{N-2}}{\Delta x^2} + O(\Delta x^2).$$

Another way is to change the mesh:

$$x_N = 1 + 1/2\Delta x, \quad N\Delta x = 1 + 1/2\Delta x.$$

Thus, the first order derivative can be approximated by using only two nodes:

$$\left. \frac{dC}{dx} \right|_{x=1} = \frac{C_N - C_{N-1}}{\Delta x} + O(\Delta x^2).$$

Overall, application of FD requires the computation of functions (such as a source term) only on nodal points resulting in a low computational expense (other methods, e.g. finite elements described further on, require computation of integrals of these functions). Furthermore, the application of FD to the domain of the regular shape and with uniform mesh results in the sparse and symmetrical system of linear (if the term  $f$  in (2.21) is linear) or nonlinear equations.

### 2.5.2 Finite volumes

Finite volumes (FV) is an extension of FD method (Versteeg and Malalasekera, 2007) and, at the same time, a transition point to a more general finite elements method. It has been used in models that consider the hydrology problem in heaps (Cariaga *et al.*, 2005). The idea of the method is illustrated with the one-dimensional diffusion equation (2.21).

The segment  $[0, 1]$  is meshed with the set of nodal points  $x_1, x_2, \dots, x_N$  (they do not need to be distributed uniformly). Each nodal point  $x_i$  is the centre of the *control volume*, faces (boundaries) of this control volume are chosen to be midway between nearest nodal points, i.e. for the nodal point  $x_i$  the control volume is the segment

$$\left[ \frac{x_{i-1} + x_i}{2}, \frac{x_i + x_{i+1}}{2} \right].$$

The left boundary of the segment will be denoted by  $x_i^w$  while the right one by  $x_i^e$  (West and East).

Equation (2.21) is integrated over each control volume yielding the following equation (Versteeg and Malalasekera, 2007):

$$\int_{x_i^w}^{x_i^e} \frac{d}{dx} \left( D \frac{dC}{dx} \right) dx + \int_{x_i^w}^{x_i^e} f dx = D \frac{dC}{dx} \Big|_{x_i^w}^{x_i^e} + \bar{f}_i (x_i^e - x_i^w) = 0, \quad (2.26)$$

where  $\bar{f}$  is the average value of function  $f$  over the control volume. The first term of the equation expresses flux through the boundaries of the control volume. In the simplest case,  $C(x)$  is assumed to be a constant at each control volume, i.e. it is approximated by the set of values  $\{C_1, C_2, \dots, C_N\}$ , where  $C_i = C(x_i)$ . Thus, the flux is evaluated at volume faces using values of concentration  $C$  at nodal points:

$$\frac{dC}{dx} \Big|_{x_i^w} = \frac{C_i - C_{i-1}}{\Delta x_i} + O(\Delta x_i^2), \quad (2.27)$$

$$\frac{dC}{dx} \Big|_{x_i^e} = \frac{C_{i+1} - C_i}{\Delta x_{i+1}} + O(\Delta x_{i+1}^2), \quad (2.28)$$

where  $\Delta x_i$  denotes the distance between  $x_{i-1}$  and  $x_i$  points. These expressions are second order approximations since both derivatives are calculated using central differences.

In most practical cases, function  $f$  depends on the concentration  $C$  among other parameters, implying that  $\bar{f}_i$  depends on  $C$  as well. Normal practise in FV method is to approximate this dependence using the linear expression:

$$\bar{f}_i = f_i^* + \Phi_i C_i, \quad (2.29)$$

where  $\Phi_i = \partial \bar{f}_i / \partial C_i$ . Substitution of this expression with (2.27) and (2.28) into equation (2.26) leads to the linear set of equations:

$$-D \frac{C_i - C_{i-1}}{\Delta x_i} + D \frac{C_{i+1} - C_i}{\Delta x_{i+1}} + f_i^* + \Phi_i C_i = 0 \quad (2.30)$$

To complete the system, BCs must be applied. Boundary condition (2.22) can be applied if the first nodal point is  $x_0 = 0$ , then the concentration on the control volume  $[0, x_1/2]$  is stated to be equal to  $C^*$ . Boundary condition (2.23) is applied naturally for the last nodal point  $x_N$ , by stating the flux through the face  $x_N^e = 1$  to be equal to  $\nu$ .

Overall, the finite volume method offers a sophisticated approach to approximate a spatial derivative. It can handle domains with complex shapes (the shape of domain is implied through the calculation of the volume) and is not restricted to a uniform mesh. However, it is still difficult to obtain a higher order of approximation, since schemes that use higher resolution methods are usually unstable (Bakhvalov *et al.*, 2006).

### 2.5.3 Finite elements

A more comprehensive numerical approach solve PDEs on complex domains with a higher order of approximation is the finite elements (FE) method. The advantages of FE come with the price of higher computational costs. FE methods have been used widely in the theory of elasticity and computational fluid dynamics (Fish and Belytschko, 2007) but have been overlooked for heap leach modelling so far, although there are models exploiting it (for example, Sidborn *et al.*, 2003, by using a commercial package). The idea of the method is illustrated on a generalised form of equation (2.21). Using  $\nabla$ -notation it can be rewritten in the form:

$$\nabla \cdot \mathbf{D} \nabla C + f = 0, \quad (2.31)$$

where  $\mathbf{D}$  is a second-order tensor (i.e. can be represented as a matrix  $\{D_{ij}\}$ , each element of this matrix can be a function of spatial variables). Boundary condition (2.20) in this notation has the form:

$$-\mathbf{D} \nabla C \Big|_{\mathbf{x} \in \partial\Omega_2} = \boldsymbol{\nu}, \quad (2.32)$$

where  $\mathbf{x} = (x, y, z)$  denotes a radius-vector in  $\mathbb{R}^3$ .

The solution of equation (2.31),  $C(\mathbf{x})$ , lies in the space of functions that have continuous second derivatives on the domain  $\mathbb{C}^2(\Omega)$ . If the inner product (and derived norm) on this space is defined as

$$u \cdot v = \int_{\Omega} uv \, d\sigma,$$

it is known that this space is incomplete (Reddy, 2007), i.e. the fundamental (Cauchy) series  $\{u_n\}$ :

$$\lim_{m,n \rightarrow \infty} \|u_m - u_n\| = 0$$

does not always have a limit in this space.

The first step is to replace this space by another one, where the solution can be found more easily. This can be achieved by constructing a weak form of the equation (2.31):

- The  $\mathbb{C}^2(\Omega)$  space for function  $C(\mathbf{x})$  is replaced by the set of functions the squares of which can be integrated with the squares of their first derivatives and that have  $C^*$  value on the boundary  $\partial\Omega_1$  (Hughes, 1987):

$$\mathcal{S}_{C^*}(\Omega) = \left\{ f(\mathbf{x}) \in L_2(\Omega) \left| \int_{\Omega} (f^2 + (\nabla f)^2) d\sigma < \infty, f|_{\partial\Omega_1} = C^* \right. \right\}.$$

Additionally, a linear space

$$\mathcal{S}_0(\Omega) = \left\{ f(\mathbf{x}) \in L_2(\Omega) \left| \int_{\Omega} (f^2 + (\nabla f)^2) d\sigma < \infty, f|_{\partial\Omega_1} = 0 \right. \right\} \quad (2.33)$$

is introduced, which is similar to the set  $\mathcal{S}_{C^*}(\Omega)$ , with the only difference that functions from the the former equal to zero on the boundary  $\partial\Omega_1$ .

- Equation (2.31) is multiplied by a test function  $w(\mathbf{x}) \in \mathcal{S}_0(\Omega)$  and integrated over the space (applying Green's formula (Fish and Belytschko, 2007) and boundary conditions):

$$\int_{\Omega} \mathbf{D} \nabla C \cdot \nabla w d\sigma + \int_{\partial\Omega_2} \boldsymbol{\nu} \cdot \mathbf{n} w ds - \int_{\Omega} f w d\sigma = 0, \quad (2.34)$$

where  $\mathbf{n}$  is the normal vector to the boundary of the domain.

The next step is discretisation of the equation (2.34). Domain  $\Omega$  is meshed to sub-domains with some typical length  $h$ . The classical choice for each sub-domain is a tetrahedron, however some methods use hexahedra. Moreover, for higher degrees of approximation, edges and faces of these polyhedra can be bent. *Shape functions*  $\psi_{ij} \in \mathcal{S}_0(\Omega)$  are chosen on each sub-domain  $\Delta\Omega_i$ . They are chosen to be independent on this sub-domain and to be identical to zero outside the sub-domain. In addition, these shape functions between adjacent sub-domains must join into basis functions  $\varphi_i \in \mathcal{S}_0(\Omega)$ .

This can be illustrated using a one-dimensional example. The whole domain (the closure of the domain) is the segment  $[0, 1]$ , which is divided into sub-segments (not necessarily

equal). On each sub-segment (cell  $i$ )  $[x_{i-1}, x_i]$  piecewise linear shape functions

$$\begin{aligned}\psi_{i-1,1}(x) &= \frac{x - x_i}{x_{i-1} - x_i}, & x \in [x_{i-1}, x_i] \\ \psi_{i-1,1}(x) &= 0, & x \notin [x_{i-1}, x_i], \\ \psi_{i-1,2}(x) &= \frac{x - x_{i-1}}{x_i - x_{i-1}}, & x \in [x_{i-1}, x_i] \\ \psi_{i-1,2}(x) &= 0, & x \notin [x_{i-1}, x_i]\end{aligned}$$

are chosen. The basis function  $\varphi_{i-1}$  is constructed as a sum of two shape functions  $\psi_{i-1,2}$  and  $\psi_{i-1,1}$ , which equals to zero outside of the segment  $[x_{i-2}, x_i]$ . Similarly, quadratic shape functions can be chosen, leading to higher order of approximation:

$$\begin{aligned}\psi_{i,1} &= a_1(x - x_i^c)(x - x_i), \\ \psi_{i,2} &= a_2(x - x_{i-1})(x - x_i), \\ \psi_{i,3} &= a_3(x - x_{i-1})(x - x_i^c),\end{aligned}$$

where  $x_i^c$  is the centre of the cell  $i$  and coefficients  $a_j$  are chosen such, that these functions equal to one at  $x_{i-1}$ ,  $x_i^c$  and  $x_i$  respectively (and  $\psi_{ij} = 0$  outside of  $[x_{i-1}, x_i]$ ). Shape functions  $\psi_{i,2}$  form one sub-set of basis functions, while the sum of two functions  $\psi_{i-1,3}$  and  $\psi_{i,1}$  form another sub-set, resulting in some basis functions being associated with cells  $[x_{i-1}, x_i]$  while others with nodal points  $x_i$ . However, since quadratic functions can have negative values, they can be unsuitable for some applications.

To continue a general case, the sub-space  $\mathcal{S}_0^h(\Omega) \subset \mathcal{S}_0(\Omega)$  is formed as a linear span of basis functions  $\varphi_i$ , i.e. each function  $g(x)$  in this space has a form

$$g(x) = \sum_i g_i \varphi_i(x).$$

The approximate solution of equation (2.34) is constructed as follows (Hughes, 1987):

$$C^h(x) = \overline{C}^*(x) + C_0^h(x) = \overline{C}^*(x) + \sum_j C_j \varphi_j(x), \quad (2.35)$$

where  $\overline{C}^*(x) \in \mathcal{S}_{C^*}$  is some fixed function and  $C_0^h(x) \in \mathcal{S}_0^h$  (Bubnov-Galerkin method). The choice of  $w(x) = \varphi_i(x)$  and the substitution of (2.35) into equation (2.34) leads to the



following expression:

$$\sum_j C_j \int_{\Omega} \mathbf{D} \nabla \varphi_j \cdot \nabla \varphi_i d\sigma + \int_{\partial\Omega_2} \nu \varphi_i ds - \int_{\Omega} f \varphi_i d\sigma + \int_{\Omega} \mathbf{D} \nabla \bar{C}^* \cdot \nabla \varphi_i d\sigma = 0. \quad (2.36)$$

This equation can be written in the compact form, if the following notation is introduced:

$$K_{ij} = \int_{\Omega} \mathbf{D} \nabla \varphi_j \cdot \nabla \varphi_i d\sigma,$$

$$F_i = - \int_{\Omega} f \varphi_i d\sigma + \int_{\partial\Omega_2} \nu \varphi_i ds + \int_{\Omega} \mathbf{D} \nabla \bar{C}^* \cdot \nabla \varphi_i d\sigma. \quad (2.37)$$

Then the vector form of equation (2.36) is as follows:

$$\mathbf{K} \mathbf{C} + \mathbf{F} = 0, \quad (2.38)$$

i.e. it is a set of algebraic equations.

The development of the scheme for the arbitrary domain shows that the FE method can be easily applied to domains with complex shapes. Additionally, there is no restriction for the regularity (i.e. sub-domains do not have to be equal) of the mesh. Higher degrees of approximation can be achieved either by re-meshing the grid or by increasing the order of shape functions. The trade-off for such flexibility is the need for additional calculations, particularly, the evaluation of integrals in order to construct a system matrix for equation (2.38).

## 2.5.4 Time discretisation

Equation (2.17) constitutes a non-steady state problem. Thus, the integration over the time needs to be performed resulting in the development of the scheme for time discretisation.

There are two main ways to deal with the first order time derivatives: explicit and implicit schemes. Explicit schemes use the information about function values from previous time steps. Thus, the calculation for the current step is straightforward. For example, the easiest (explicit) forward Euler method for the equation:

$$\frac{dx}{dt} = f(x, t) \quad (2.39)$$



takes the following form:

$$\frac{x^{k+1} - x^k}{\Delta t} = f(x^k, t^k),$$

where  $\Delta t$  denotes the time step and  $k$  is the time domain counter, i.e.  $t^k = \Delta t k$  (if the time step is kept as a constant). The solution of this equation for the time step  $k + 1$  can be found from the expression:

$$x^{k+1} = x^k + \Delta t f(x^k, t^k).$$

The forward Euler method guarantees only the first order approximation (Bakhvalov *et al.*, 2006), but there are other schemes with higher orders. However, they all have one major disadvantage: they are not absolutely stable, which means that the error between the true solution and its approximation can be unbound for some values of  $\Delta t$ , i.e. solution can be meaningful only if  $\Delta t$  is restricted to some values. For example, the forward Euler method is stable only if (Chizhonkov, 2006)

$$\Delta t < \frac{2}{|\partial f_i / \partial x_j|}.$$

Thus, if  $f(x, t)$  changes sharply with the the change of  $x$ , this scheme requires  $\Delta t$  to be very small to ensure stability.

In contrast, some implicit methods provide absolute stability. For example, the (implicit) backward Euler method for equation (2.39) is defined as follows:

$$\frac{x^{k+1} - x^k}{\Delta t} = f(x^{k+1}, t^{k+1}).$$

This method is proved to be absolutely stable, i.e. it does not impose any restriction on the value of  $\Delta t$  (Bakhvalov *et al.*, 2006). The trade-off is the need to solve non-linear equation, since the solution  $x^{k+1}$  has been used to calculate right-hand-side function. As the forward Euler method, the implicit one guarantees only the first order approximation. If the right-hand-side part of the equation is slightly modified:

$$\frac{x^{k+1} - x^k}{\Delta t} = \frac{1}{2} (f(x^{k+1}, t^{k+1}) + f(x^k, t^k)),$$

then this method ensures the second order approximation as well as absolute stability (Crank-Nicolson method). However, not all implicit methods are absolutely stable, for example, the

multi-step Adams method for order higher than two is not absolutely stable (Chizhonkov, 2006).

There is no evidence in the open literature that any explicit method has been used for heap leach modelling. It can be explained by the fact, that problems that involve chemical reaction effects tend to be stiff and, thus, explicit algorithms to be unstable. Therefore, the choice of absolutely stable methods (even if they require more computations and have lower approximation order) is sensible. For example, Dixon and Hendrix (1993a) and Sheikhzadeh *et al.* (2005) have used the backward Euler method, whereas HeapSim has used the Crank-Nicolson method (Petersen and Dixon, 2005). Ogbonna (2006) has used one of Adams's methods, but a particular method has been automatically chosen by the external library, SciPy, used in that work, so it remains unclear, if the order higher than two has been used (i.e. not absolutely stable) or only up-to the second order method has been chosen.

### 2.5.5 Solution of non-linear algebraic equations

The application of an implicit time discretisation scheme, or space discretisation method, for steady-state problem results in a non-linear algebraic equation (or set of equations).

A set of algebraic equations can be stated in a vector form as  $\mathbf{F}(\mathbf{x}) = 0$ . A general numerical method to solve this equation is to construct the equivalent equation  $\mathbf{x} = \mathbf{G}(\mathbf{x})$  such that function  $\mathbf{G}(\mathbf{x})$  is a contraction mapping, i.e. for two different vectors  $\mathbf{x}$  and  $\mathbf{y}$

$$\|\mathbf{G}(\mathbf{x}) - \mathbf{G}(\mathbf{y})\| \leq \kappa \|\mathbf{x} - \mathbf{y}\| \quad (2.40)$$

for some  $\kappa < 1$  (fixed point method). Thus, the equation  $\mathbf{x} = \mathbf{G}(\mathbf{x})$  has a unique solution which is identical to the solution of the initial set of equations (Bakhvalov *et al.*, 2006).

The application of the fixed point method can be demonstrated on equation (2.38):

$$\mathbf{C} = -\mathbf{K}^{-1} \mathbf{F}(\mathbf{C}),$$

where  $\mathbf{F}$  is a non-linear function of  $\mathbf{C}$ . Numerical solution is constructed using iterations:

$$\mathbf{C}^{(n+1)} = -\mathbf{K}^{-1} \mathbf{F}(\mathbf{C}^{(n)}) \quad (2.41)$$

with some initial approximation  $\mathbf{C}^{(0)}$ . For a non-linear function  $\mathbf{F}(\mathbf{C})$  it is not always possible to verify condition (2.40) *a priori*. Moreover,  $\mathbf{F}(\mathbf{C})$  can be contraction mapping only

on a certain subspace of the total space of  $C$ . Thus, the solution of non-linear equations by using fixed point method implies practical difficulties.

A more complex application of the general fixed point method is done in Newton's method. The method can be illustrated on a general example  $F(x) = 0$ . Iterations are constructed using the expression:

$$x^{(n+1)} = x^{(n)} - J^{-1}(x^{(n)})F(x^{(n)}), \quad (2.42)$$

where matrix  $J$  is a Jacobian matrix of the system:

$$J_{ij} = \frac{\partial F_i}{\partial x_j}.$$

The advantage of the method is fast convergence (Bakhvalov *et al.*, 2006). However, this method is associated with high computational expense as for the set of  $N$  equations and  $N$  variables, it requires the computation of  $N^2$  derivatives at each step. To simplify computations, evaluation of matrix  $J$  can be performed only at initial point  $x^{(0)}$ .

### 2.5.6 Solution of sparse system of linear equations

Linear systems usually appear in the process of solution of non-linear equations (2.41) and (2.42). If the system matrix is the result of the application of a space discretisation scheme, such as FD (2.25) or FE (2.38), then this matrix is sparse. Therefore, special methods, which can help to keep low computational cost, can be applied.

The usual way to deal with sparse linear systems

$$Ax = b$$

is to construct iterative approximations using the following structure:

$$x^{(n+1)} = Bx^{(n)} + c.$$

Any particular method is defined by the choice of matrix  $B$  and vector  $c$ . These methods can be divided into two groups: stationary methods, where neither  $B$  nor  $c$  depend on the iteration count  $n$ , and non-stationary methods (Barrett *et al.*, 1993).

*Jacobi method* is an example of the stationary method. The  $i$ th equation of the system looks as follows:

$$\sum_j a_{ij}x_j = b_i.$$

Component  $x_i$  can be expressed through others:

$$x_i = \frac{\left(b_i - \sum_{j \neq i} a_{ij}x_j\right)}{a_{ii}}.$$

Thus, the iterative scheme

$$x_i^{(n+1)} = \frac{\left(b_i - \sum_{j \neq i} a_{ij}x_j^{(n)}\right)}{a_{ii}}$$

can be used. Or in matrix term it can be expressed as follows:

$$\mathbf{x}^{(n+1)} = -\mathbf{D}^{-1}(\mathbf{L} + \mathbf{U})\mathbf{x}^{(n)} + \mathbf{D}^{-1}\mathbf{b},$$

where  $\mathbf{D}$ ,  $\mathbf{L}$  and  $\mathbf{U}$  represent diagonal, strictly low-triangle and upper-triangle parts of matrix  $\mathbf{A}$  (Barrett *et al.*, 1993).

The *conjugate gradient* (CG) method is one of the oldest non-stationary methods. It is effective for positively defined symmetrical matrices  $\mathbf{A}$ . The idea of the method is to minimise the quadratic function

$$f(\mathbf{x}) = \frac{1}{2}\mathbf{x}^T\mathbf{A}\mathbf{x} - \mathbf{b}\mathbf{x},$$

which is minimized when its gradient

$$\nabla f = \mathbf{A}\mathbf{x} - \mathbf{b} = 0.$$

The method is based on the construction of a series of three vectors: approximate solution  $\mathbf{x}^{(n)}$ , residual  $\mathbf{r}^{(n)}$  and search direction  $\mathbf{p}^{(n)}$ . The next approximation is found, providing that the next residual is orthogonal to the previous one (it can be shown, that it is orthogonal to all previous residuals). Thus, after  $N$  steps (where  $N$  is the size of the system) this method finds the exact solution. Formulae to find next approximation are the following:

$$\begin{aligned}\mathbf{x}^{(n+1)} &= \mathbf{x}^{(n)} + \alpha_{n+1}\mathbf{p}^{(n+1)}, \\ \mathbf{r}^{(n+1)} &= \mathbf{r}^{(n)} - \alpha_{n+1}\mathbf{A}\mathbf{p}^{(n+1)}, \\ \mathbf{p}^{(n+1)} &= \mathbf{r}^{(n)} + \beta_n\mathbf{p}^{(n)},\end{aligned}$$

where the choice of  $\alpha_{n+1}$ :

$$\alpha_{n+1} = \frac{\mathbf{r}^{(n)T} \mathbf{r}^{(n)}}{\mathbf{p}^{(n+1)T} \mathbf{A} \mathbf{p}^{(n+1)}}$$

minimises the residual in the norm  $\mathbf{r}^{(n+1)T} \mathbf{A} \mathbf{r}^{(n+1)}$ , whereas the choice of  $\beta_n$ :

$$\beta_n = \frac{\mathbf{r}^{(n)T} \mathbf{r}^{(n)}}{\mathbf{r}^{(n-1)T} \mathbf{r}^{(n-1)}}$$

ensures the orthogonality of residuals  $\mathbf{r}^{(n)}$  and  $\mathbf{r}^{(n+1)}$  (Barrett *et al.*, 1993). As it can be seen, to make the transition from the  $n$ -th to the  $(n+1)$ -th iteration, only four vectors  $\mathbf{x}^{(n)}$ ,  $\mathbf{r}^{(n-1)}$ ,  $\mathbf{r}^{(n)}$ , and  $\mathbf{p}^{(n)}$  need to be kept in the memory, which is a very small number in comparison with some other methods. For example, the use of memory in the generalised minimal residual method (GMRES) increases linearly with the increase of iterative count  $n$ .

GMRES, in contrast to CG, can work with non-symmetrical systems. One of the alternative methods to solve such equations and keep low computational expense is the *bi-conjugate gradient method* (BiCG). BiCG constructs two mutually orthogonal sequences of residual vectors  $\mathbf{r}^{(n)}$  and  $\tilde{\mathbf{r}}^{(n)}$  but does not provide the minimisation of residual at each step. The method is summarised with formulae:

$$\mathbf{r}^{(n+1)} = \mathbf{r}^{(n)} - \alpha_{n+1} \mathbf{A} \mathbf{p}^{(n+1)} \quad \text{and} \quad \tilde{\mathbf{r}}^{(n+1)} = \tilde{\mathbf{r}}^{(n)} - \alpha_{n+1} \mathbf{A}^T \tilde{\mathbf{p}}^{(n+1)},$$

where  $\mathbf{p}^{(n+1)}$  and  $\tilde{\mathbf{p}}^{(n+1)}$  are search directions defined as follows:

$$\mathbf{p}^{(n+1)} = \mathbf{r}^{(n)} + \beta_n \mathbf{p}^{(n)} \quad \text{and} \quad \tilde{\mathbf{p}}^{(n+1)} = \tilde{\mathbf{r}}^{(n)} + \beta_n \tilde{\mathbf{p}}^{(n)}.$$

The choice of coefficient  $\alpha_{n+1}$  and  $\beta_n$ :

$$\alpha_{n+1} = \frac{\tilde{\mathbf{r}}^{(n)T} \mathbf{r}^{(n)}}{\tilde{\mathbf{p}}^{(n+1)T} \mathbf{A} \mathbf{p}^{(n+1)}} \quad \text{and} \quad \beta_n = \frac{\tilde{\mathbf{r}}^{(n)T} \mathbf{r}^{(n)}}{\tilde{\mathbf{r}}^{(n-1)T} \mathbf{r}^{(n-1)}}$$

ensures that the sequence of residuals is bi-orthogonal:

$$\tilde{\mathbf{r}}^{(j)T} \mathbf{r}^{(i)} = 0 \quad \text{if} \quad i \neq j.$$

The method starts with the initial guess  $\mathbf{x}^{(0)}$ , the initial residual is calculated directly:

$$\mathbf{r}^{(0)} = \mathbf{b} - \mathbf{A} \mathbf{x}^{(0)}$$

and the vector  $\tilde{\mathbf{r}}^{(0)}$  is chosen to be equal to  $\mathbf{r}^{(0)}$ . The next approximation of the solution is calculated in the same way as for the CG method.

BiCG has been reported to converge rapidly but not consistently, i.e. there are some cases where the method can break down. If the system is such that this method fails, it either can be restarted at the step before the break (however, this does not guarantee that it will finally converge) or it can be switched to the more robust GMRES (Barrett *et al.*, 1993).

In addition, both CG and BiCG can be used with a preconditioner to facilitate convergence: instead of solving system  $\mathbf{Ax} = \mathbf{b}$  a preconditioned method solves

$$\tilde{\mathbf{A}}^{-1} \mathbf{Ax} = \tilde{\mathbf{A}}^{-1} \mathbf{b},$$

where matrix  $\tilde{\mathbf{A}}$  is close to  $\mathbf{A}$ . In the simplest case, it can be chosen as a diagonal part of  $\mathbf{A}$ . Thus, its inverse  $\tilde{\mathbf{A}}^{-1}$  can be calculated easily.

### 2.5.7 Minimisation of functions

Model calibration involves comparison with experimental data and adjusting some model parameters to fit this data. This task can be formalised as follows: by varying parameters

$$(\alpha_1, \alpha_2, \dots, \alpha_P) \in K^P$$

to minimise a function

$$f(\alpha_1, \alpha_2, \dots, \alpha_P): K^P \rightarrow \mathbb{R}$$

that represents a difference between a model prediction and the experimental data.

The standard way to minimise a function  $f$  is to determine the direction that leads to a decrease of its value, take this direction and repeat the procedure (Press *et al.*, 2002). Methods vary in the way how they find this direction. The major problem with these methods is that function  $f$  can have local minima. In this case it is highly likely that the procedure would result in one of these local minima and would not find the global one.

The alternative approach is to randomly search the whole space  $K^P$  to coarsely determine the position of the global minimum, which is followed by more refine search near it. The idea can be illustrated with the *simulated annealing method* (Press *et al.*, 2002).

The minimisation starts at some initial guessed value  $\alpha = (\alpha_1, \dots, \alpha_P)$ . A new value  $\alpha'$  is calculated on the base of the initial guess by random change of values  $\alpha_p$ . If  $\alpha'$  decreases the value of  $f$ :

$$\Delta f = f(\alpha') - f(\alpha) < 0$$

it is taken as a new guess. However, if  $\Delta f > 0$  then  $\alpha'$  can be taken as a new guess with the probability:

$$\text{Prob} = \exp(-\Delta f/T),$$

where  $T$  is a parameter of the minimisation method. Notice that if  $\Delta f < 0$  the probability is greater than one, therefore,  $\alpha'$  is always taken as a new guess in this case.

The crucial part of the method is a “cooling schedule”: as the minimisation procedure progresses the value of  $T$  is decreasing, leading to the decrease in the probability that a new value  $\alpha'$  that results in  $\Delta f > 0$  would be taken. Thus, initially this method allows to go over local maxima of  $f$  in order to find a global minimum. However, later in the procedure, it will almost always decrease the value of  $f$ .

The simulated annealing method has been applied successfully to many mathematical problems, especially discrete ones (such as the travelling salesperson problem) where classical methods cannot be applied (Press *et al.*, 2002).

## 2.6 Techniques of programme development

The implementation of models evolved with time. Initially, models, especially developed before the 1990's, used FORTRAN language (Petersen and Dixon, 2005) as it was common practice for engineering applications. Later models often opted for commercial packages (Bennett *et al.*, 2003; Sidborn *et al.*, 2003). The aim of this section is to briefly review different programming approaches and how they can be applied for modelling of heap systems.

Older models, mostly written in FORTRAN or Pascal, used one form of procedural programming. This paradigm states that the programme needs to be broken into the set of procedures (functions), which implement particular functionality that can be used as a part of



the main programme algorithm. FORTRAN77, Pascal, C and other programming languages directly support this paradigm.

Procedures can be grouped into different modules, leading to modular programming. The main difference between the former and the latter is that modular programming is mainly focused on the organisation of the data on which programmes operate, so modules usually contain not only procedures, but data or definitions for data organisation (for example, records in terms of Pascal, or structures in C). Some languages, such as Pascal or FORTRAN9x, have extensive support of modular programming, while others (for example, FORTRAN77) have very limited support.

Object oriented programming (OOP) shifts the focus on data even more, introducing abstract type definitions, re-use of code by inheritance and polymorphism (Stroustrup, 1997). In OOP data and functions, that operate on the data, are merged together. OOP has become an upstream development technique from the early 1990s, especially due to the use of OOP for developing programmes with an interactive user interface. Modern widely used object-oriented programming languages include C++ and Java.

Functional programming has been mainly used for mathematical research, especially in the area of discrete mathematics (Lisp, a list processor, is regarded as the first functional language). However, this approach has gained popularity recently since it offers some advantages over other paradigms. For example, concurrency, implemented using functional programming, has been proved to be more robust and less prone to hidden errors than OOP (Wampler and Payne, 2009). The main idea of this approach is to make all data immutable (wherever possible), if it needs to be changed, new data must be generated. This implies that if any function is called with the same argument it must return the same result. This idea appears to solve many problems that traditional approaches have, for example, data synchronisation between different parts of the programme. The main drawback of the functional programming is the extensive use of the memory and a computational cost associated with it.

Overall, different paradigms offer different advantages (Stroustrup, 1997). OOP can reduce the amount of time spent on programme developing due to source code re-use and polymorphic behaviour, but the resulting programme might have slower performance. The



partial use of functional programming can increase the robustness of the programme while keeping additional computational expense minimal.

## 2.7 Closure and objectives

The present review shows gaps in two different areas:

- On the one hand, there is a limited understanding of the chloride assisted leaching of chalcopyrite in the presence of mass transfer constraints.
- On the other hand, there are a number of limitations in heap leach modelling, such as a lack of generic properties and systematic calibration and validation.

This project aims to investigate the effect of mass transfer constraints on the chloride leaching of chalcopyrite using modelling as a research tool. Thus, it targets both areas.

The first step to be taken is to obtain more information from the experimental data presented in section 2.3.2. The main goal of this step is to estimate the rate of leaching and its limiting factors.

In parallel, a generic agglomerate-scale model needs to be developed and proved to be robust for further use. This model then can be applied to the chloride leaching by using information obtained at the first step. Hence, this model will be able to serve as a guidance for further laboratory studies.

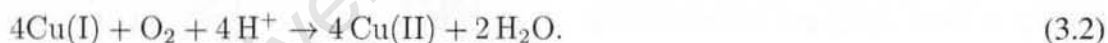
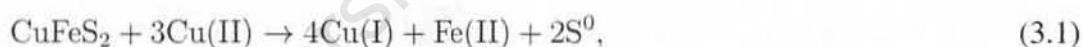
Overall, the present work merges together two topics from different areas, but that appeared to be deeply inter-connected. It aims to gain more understanding in the first area by developing and applying advanced techniques from the second one.

## Chapter 3

### Chloride Leaching Model

Chloride heap leaching is an alternative to the bioleaching to leach copper from sulphide minerals in cases when the latter cannot be used. This technology is still developing and very little research has been conducted so far.

It is still speculative what the actual reaction network for the chloride leaching of chalcopyrite is. The latest understanding can be summarised with two reactions:



Each of these reaction is, in fact, a chain of other reactions with some intermediate products (see Nicol *et al.*, 2010). However, since very little is known about the rates of those elementary reactions, at this moment, it is sensible to focus on summarised reactions (3.1) and (3.2). It should be noted, that chloride ions do not participate in the leaching directly, but are used to stabilise cuprous ions,  $\text{Cu}^+$ , in solution as Cl-complexes.

Although the experiments, conducted in stirred tank reactors, produced promising results, the results of the first attempts of column tests were comparatively poor (Petersen, 2008). A modelling approach is employed here to determine the key factor that limits the overall process.

	Bulk	Pore Lengths		
		2 mm	5 mm	10 mm
Redox Potential	682 mV	647 mV	634 mV	622 mV

Table 3.1: Basic experiment results: potential values

### 3.1 Experimental data analysis

The experiment on mass transfer constrains, described in section 2.3.2, has generated a somewhat limited amount of data (table 3.1, which is a copy of table 2.1, provided here for the convenience). However, application of a simple modelling approach can provide more detailed information for a more complex model, such as validate initial assumptions and provide estimates of leaching rates. This section describes the development of a linear microscale model, which is based on a number of assumptions to provide initial insight into the process dynamics.

The direct experimental measurements shown in table 3.1 indicate a drop in the potential value as the length of the capillary increases. Thus, they suggest an increase in  $[Cu^+]/[Cu^{2+}]$  ratio (see equation (2.5), additionally, to simplify calculations it is assumed, that ferrous ions, generated in reaction (3.1), do not affect the potential due to their low concentration). This increase is most likely due to the increase of  $[Cu^+]$  formed by reaction (3.1). This analysis aims to quantify this increase and find an estimate of the leaching rate.

First, potential data need to be mapped to Cu(I) and Cu(II) concentrations. The Nernst equation (2.5) provides the means to do this. However, in the strong chloride environment of the experiments these ions tend to form stable complexes with chloride ions. Hence,  $[Cu^+]$  and  $[Cu^{2+}]$  do not account for the total concentration of Cu(I) and Cu(II) respectively. The correlation can be calculated based on thermodynamic equations (M. Nicol, personal communication, 2010):

$$[Cu^+] = \frac{[Cu(I)]}{1 + K'_1[Cl^-] + K_1K_2[Cl^-]^2 + K_1K_2K_3[Cl^-]^3} \quad (3.3)$$

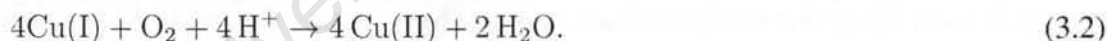
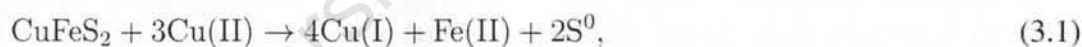
$$[Cl^-] = [Cl] - [Cu^+](K_1[Cl^-] + 2K_1K_2[Cl^-]^2 + 3K_1K_2K_3[Cl^-]^3) \quad (3.4)$$

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$$[Cl^-] = [Cl] - [Cu^+](K_1[Cl^-] + 2K_1K_2[Cl^-]^2 + 3K_1K_2K_3[Cl^-]^3) \quad (3.4)$$

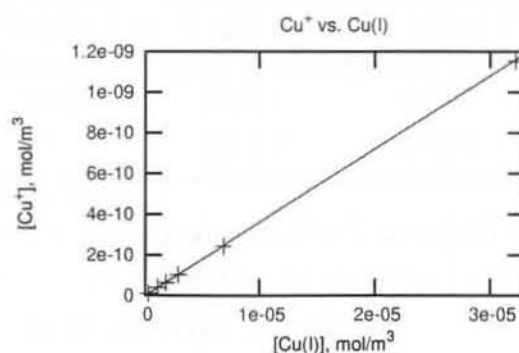


Figure 3.1: Almost linear dependence of  $[Cu^+]$  on  $[Cu(I)]$

where constants are defined as follows:  $K_1 = 10^{2.7}$ ,  $K_1K_2 = 10^{5.8}$ ,  $K_1K_2K_3 = 10^{5.52}$ . Similarly, total Cu(II) concentration can be found from equations:

$$[Cu^{2+}] = \frac{[Cu(II)]}{1 + K'_1[Cl^-]} \quad (3.5)$$

$$[Cl^-] = [Cl] - K'_1[Cl^-][Cu^{2+}] \quad (3.6)$$

where constant  $K'_1 = 10^{0.14} \approx 1.38038$ .

Although the set of equations (3.3) and (3.4) are non-linear, it is easy to show that for  $[Cl] = 0.2 \text{ mol/L}$  at  $25^\circ\text{C}$  and the range of  $[Cu(I)]$  between  $2.8 \times 10^{-7}$  and  $3.2 \times 10^{-5} \text{ mol/L}$ ,  $Cu^+$  depends on Cu(I) almost linearly:

$$[Cu^+] = \lambda[Cu(I)] \quad (3.7)$$

with  $\lambda \approx 3.6 \times 10^{-5}$  (see figure 3.1 for the plot of  $Cu^+$  vs.  $Cu(I)$ ).

A simple linear model can be formulated to estimate leaching reaction rate. In this model it is assumed that there is no significant oxidation of Cu(I), i.e. reaction (3.1) is the only reaction that occurs in the system.

Although this assumption is quite speculative (a more comprehensive model, developed in the next section, does not require it), it enables very transparent analysis of the experimental data. Under this assumption, the concentrations of Cu(I) and Cu(II) change linearly

Species	Bulk	2 mm	5 mm	10 mm	Unit
Cu(I)	0.28070	1.0961	1.8178	2.8996	$\times 10^{-3}$ mol/m <sup>3</sup>
Cu(II)	7.8737	7.8731	7.8726	7.8718	mol/m <sup>3</sup>

Table 3.2: Concentrations of Cu(I) and Cu(II) in bulk solution and at the mineral surface for different capillaries

through the capillary. Therefore, fluxes ( $\mathcal{N}$ ) can be determined by linear expressions:

$$\mathcal{N}(\text{Cu(I)}) = -D([\text{Cu(I)}] - [\text{Cu(I)}]_{\text{bulk}})/l_{\text{max}}, \quad (3.8)$$

$$\mathcal{N}(\text{Cu(II)}) = -D([\text{Cu(II)}] - [\text{Cu(II)}]_{\text{bulk}})/l_{\text{max}}, \quad (3.9)$$

$$(3.10)$$

where  $[\text{Cu(I)}]$  and  $[\text{Cu(II)}]$  are concentrations at the mineral surface and  $l_{\text{max}}$  is the length of the capillary. Due to reaction stoichiometry, these fluxes must satisfy a mass balance equation at the mineral surface:

$$1/4\mathcal{N}(\text{Cu(I)}) = -1/3\mathcal{N}(\text{Cu(II)}). \quad (3.11)$$

The Nernst equation (2.5), correlations (3.3), (3.4), (3.5) and (3.6), and mass balance equation (3.11) combined with fluxes definitions (3.8) and (3.9) form the complete system to find concentrations of Cu(I) and Cu(II) at the mineral surface. It should be noted that the model is not very stable, due to the nature of the inverse of the Nernst equation: a small change in the potential value will result in a large change of Cu(I) concentration (if  $[\text{Cu(II)}]$  is fixed).

The results of the application of this model are shown in table 3.2. From the results it is evident that the change of Cu(II) concentration is insignificant in comparison with the change of Cu(I) concentration. Thus, there is a significant accumulation of Cu(I) ions near the mineral surface resulting in a lower potential value. Table 3.3 shows fluxes and leaching rates ( $1/4\mathcal{N}_{\text{Cu(I)}}$ ) that have been derived based on Cu(I) concentrations ( $D = 1.2 \times 10^{-9}$  m<sup>2</sup>/sec, Moats *et al.* (2000)). The details of calculations can be found in the Appendix A.

Although the linear model enables the estimation of Cu(I) accumulation near the mineral surface, these results have been drawn under the assumption of the absence of Cu(I)

oxidation. To establish the role of the oxygen in this process, a more sophisticated model is required. This model is developed in the next section.

## 3.2 Model description

To determine the effect of oxygen in the leaching process, a steady-state diffusion model is formulated. This model is based on several assumptions:

- The system consists of Cu(I), Cu(II) and O<sub>2</sub>. It has been hypothesised (modelling assumption) that the changes of [Cu(II)] and [O<sub>2</sub>] through the capillary are small in comparison with the change of [Cu(I)]. From table 3.2 it is evident that [Cu(II)] >> [Cu(I)]. Furthermore, [O<sub>2</sub>] can be estimated as 0.15 mol/m<sup>2</sup>, which is 100 higher than concentration of Cu(I). Thus, large variations in [Cu(II)] and [O<sub>2</sub>] are not expected.
- Since the change of concentrations of Cu(II) and O<sub>2</sub> are negligibly small, all reaction rates are assumed to be independent of them (or dependent only on their concentrations in the bulk solution).
- Ferrous ions, produced by leaching reaction (3.1), do not affect the value of the potential due to their low concentration. Indeed, if the system starts iron-free, total iron concentration in the solution is four times lower than that of Cu(I) owing to the stoichiometry of the reaction (3.1). Thus, it is assumed that ferrous ions have no significant effect on the process.

The system is governed by the transport of Cu(I) through the capillary due to diffusion:

$$D \frac{d^2 C}{dl^2} - 4r_{ox} = 0, \quad (3.12)$$

Capillary length, mm	$\mathcal{N}_{\text{Cu(I)}}$ , mol/(m <sup>2</sup> · sec)	Leaching rate, mol/(m <sup>2</sup> · sec)
2	$4.89 \times 10^{-10}$	$1.22 \times 10^{-10}$
5	$3.69 \times 10^{-10}$	$9.22 \times 10^{-11}$
10	$3.14 \times 10^{-10}$	$7.86 \times 10^{-11}$

Table 3.3: Cu(I) fluxes and reaction rates, depending on the pore length



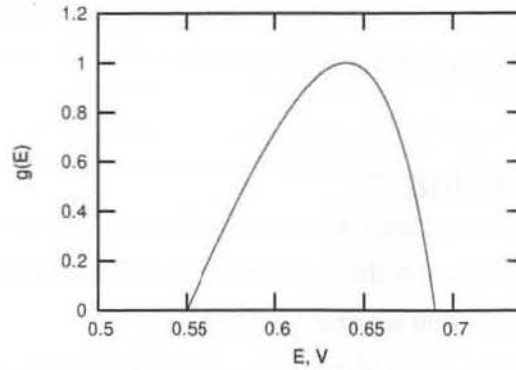


Figure 3.2: Normalised Ratkowsky function

where  $C$  is the concentration of Cu(I) and  $r_{ox}$  is the rate of Cu(I) oxidation reaction. At the mouth of the capillary the concentration is specified:

$$C \Big|_{l=0} = C^*, \quad (3.13)$$

whereas the flux is specified at the end of the capillary:

$$-D \frac{dC}{dl} \Big|_{l=l_{max}} = -4r_{cpy} \quad (3.14)$$

If the oxidation rate  $r_{ox}$  is assumed to be linear:

$$r_{ox} = k(C - C^*), \quad (3.15)$$

with the same equilibrium concentration  $C^*$  as in the bulk solution due to little change in total copper concentration, then equation (3.12) can be integrated analytically. The solution (with the applied essential boundary condition at the capillary-bulk interface) has the following form:

$$C = Be^{\sqrt{\frac{4k}{D}}l} - Be^{-\sqrt{\frac{4k}{D}}l} + C^* \quad (3.16)$$

where constant  $B$  needs to be determined by using the condition at the mineral surface.

The only information known about chalcopyrite leaching rate is that it depends on the potential value  $E$  and temperature (out of the scope of this study), and is quite insensitive to other factors, such as pH or total copper concentration (Velasquez-Yevenes *et al.*, 2010a). Thus, the rate has been stated to have the form

$$r_{cpy} = pg(E) \quad (3.17)$$

where  $g(E)$  is the function of the potential with conditions: it equals to 0 outside of the potential range of 550 mV – 700 mV and reaches the maximum value of 1 at  $E \approx 640$  mV. Since this conditions are quite loosely defined, there is a wide choice of the forms for this function. The Ratkowsky-type form has been chosen:

$$g(E) = \frac{F(E)}{F(E^{\text{opt}})},$$

where function  $F(E)$  looks as follows:

$$F(E) = (E - E^{\text{min}}) (1 - \exp(c(E - E^{\text{max}})))$$

since it allows one to specify an optimum potential by varying parameter  $c$  (the graph of  $g(E)$  is shown on figure 3.2). Function  $g(E)$ , combined with Nernst equation (2.5) and equations (3.3) and (3.4), that determines  $[\text{Cu}^+]$  on the base of  $[\text{Cu(I)}]$  defines the rate of chalcopyrite leaching:

$$r_{\text{cpy}} = p \left( g \circ E \circ \varphi \circ C(B, l_{\text{max}}) \right) \quad (3.18)$$

where  $\circ$  is a function composition and  $\varphi$  denotes a function that determines  $[\text{Cu}^+]$ :

$$[\text{Cu}^+] = \varphi([\text{Cu(I)}])$$

either by the solution of (3.3) and (3.4) or by the linear expression (3.7).

Substitution of the solution (3.16) into boundary condition (3.14) leads to the following equation:

$$B = \frac{4r_{\text{cpy}}}{D \sqrt{\frac{4k}{D}} \left( \exp \left( \sqrt{\frac{4k}{D}} l_{\text{max}} \right) + \exp \left( -\sqrt{\frac{4k}{D}} l_{\text{max}} \right) \right)} \quad (3.19)$$

where  $r_{\text{cpy}}$  depends on  $B$  through the function composition (3.18). This equation is solved numerically by the contraction mapping method (see section 2.5.5, the form of the equation enables the direct application of this method). The solution of equation (3.19) determines the profile of  $[\text{Cu(I)}]$  via equation (3.16).

$\text{Cu(II)}$  and  $\text{O}_2$  profiles can be obtained analytically once the  $\text{Cu(I)}$  profile is defined (i.e. the value of  $B$  is found). The  $\text{Cu(II)}$  concentration profile is governed by the diffusion equation:

$$D \frac{d^2 C_{\text{Cu(II)}}}{dl^2} + 4r_{\text{ox}} = 0 \quad (3.20)$$

with boundary conditions:

$$\begin{aligned} C_{\text{Cu(II)}} \Big|_{l=0} &= C_{\text{Cu(II)}}^* \\ -D \frac{dC_{\text{Cu(II)}}}{dl} \Big|_{l=l_{\max}} &= 3r_{\text{cpy}} \end{aligned}$$

where  $r_{\text{ox}}$  in this case is a known function of  $l$  (after substitution of (3.16) into (3.15)):

$$r_{\text{ox}} = k(T)B \left( e^{\sqrt{\frac{4k}{D}}l} - e^{-\sqrt{\frac{4k}{D}}l} \right)$$

Solution of equation (3.20) with applied boundary conditions has the form:

$$C_{\text{Cu(II)}} = B \left( -e^{\sqrt{\frac{4k}{D}}l} + e^{-\sqrt{\frac{4k}{D}}l} \right) + \varkappa l + C_{\text{Cu(II)}}^* \quad (3.21)$$

with  $\varkappa$  defined as follows:

$$\varkappa = \frac{1}{D} \left( -3r_{\text{cpy}} + DB \sqrt{\frac{4k}{D}} \left( e^{\sqrt{\frac{4k}{D}}l_{\max}} + e^{-\sqrt{\frac{4k}{D}}l_{\max}} \right) \right)$$

A similar equation defines the profile of oxygen concentration:

$$D_{\text{O}_2} \frac{d^2 C_{\text{O}_2}}{dl^2} - r_{\text{ox}} = 0 \quad (3.22)$$

with boundary conditions that state a constant concentration at the capillary mouth and prevent the flux at the mineral surface:

$$\begin{aligned} C_{\text{O}_2} \Big|_{l=0} &= C_{\text{O}_2}^* \\ -D_{\text{O}_2} \frac{dC_{\text{O}_2}}{dl} \Big|_{l=l_{\max}} &= 0 \end{aligned}$$

Solution of this equation has the form:

$$C_{\text{O}_2} = \frac{D}{4D_{\text{O}_2}} B \left( e^{\sqrt{\frac{4k}{D}}l} - e^{-\sqrt{\frac{4k}{D}}l} \right) + \varkappa_{\text{O}_2} l + C_{\text{O}_2}^* \quad (3.23)$$

where  $\varkappa_{\text{O}_2}$ :

$$\varkappa_{\text{O}_2} = -\frac{D}{4D_{\text{O}_2}} B \sqrt{\frac{4k}{D}} \left( e^{\sqrt{\frac{4k}{D}}l_{\max}} + e^{-\sqrt{\frac{4k}{D}}l_{\max}} \right)$$

Thus, the solution of non-linear algebraic equation (3.19) defines the profiles of all the species. Figure 3.3 shows the result of the model (source code listings can be found in Appendix A) for the parameters listed in table 3.4: most of the parameter values represent conditions of the experiments described in section 2.3.2, except the values of rate constants  $p$  and  $k$  which have been chosen arbitrarily (the model calibration with respect to these parameters is described further below). To illustrate the difference of the relative change of species concentrations, graphs 3.3 have been summarised in figure 3.4. This figure validates the assumption of the insignificant variation of  $Cu(II)$  and  $O_2$  concentrations since  $[Cu(II)]$  and  $[O_2]$  vary less than 1% whereas  $[Cu(I)]$  at the mineral surface is 8 times higher than at the capillary mouth.

### 3.3 Model calibration

Although the results from the previous section match experimental results in principle (e.g. higher  $[Cu(I)]$  at the mineral surface than at the capillary mouth), they significantly deviate from experimental data. Thus, the model needs to be calibrated and validated.

The model has several parameters with guessed values:  $k$ ,  $p$ ,  $c$ ,  $E^{\min}$ , and  $E^{\max}$ . Since there is a lack of information about the factor-function  $g(E)$ , it is sensible at this moment to assume that its parameters are suggested correctly. Therefore, the model can be calibrated by changing the reaction coefficients  $k$  and  $p$ . Additionally, this study can show the role each reaction plays in the leaching process.

The simple simulated annealing method (see section 2.5.7) has been set up to find optimum values of  $k$  and  $p$  by minimising the following functional:

$$\gamma[C_{Cu(I)}, C_{Cu(II)}] = \sqrt{\left(\frac{C_{Cu(I)}(l_{\max})}{C_{Cu(I),\exp}} - 1\right)^2 + \left(\frac{C_{Cu(II)}(l_{\max})}{C_{Cu(II),\exp}} - 1\right)^2} \quad (3.24)$$

where subscript "exp" means concentrations obtained from the experiment. It should be noted, since it is a stochastic approach, it generates some (although insignificant) variance in the final results.

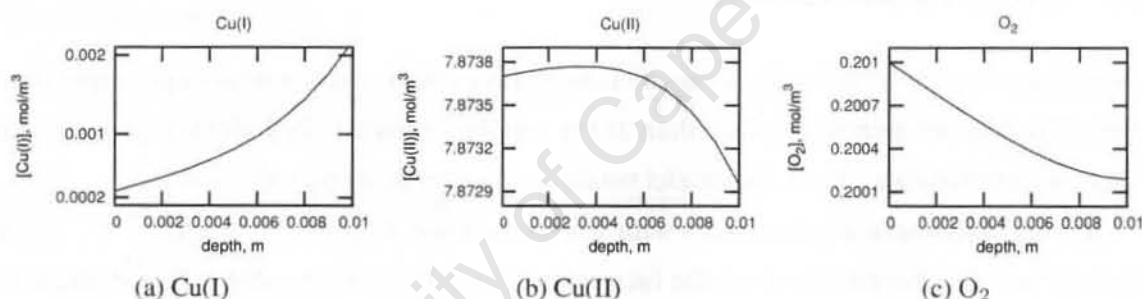
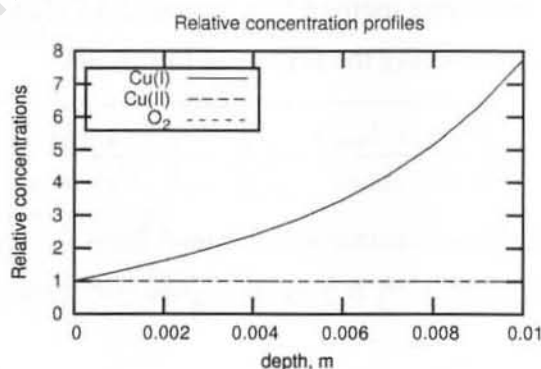
The model has been calibrated by using experimental results for the 10 mm capillary. The final values of  $\gamma$ , reaction rate constants,  $B$  and potential are listed in table 3.5 on page 55. It should be noted, that the value of  $k$  obtained from the calibration implies extremely slow

Parameter	Value	Unit	Parameter	Value	Unit
$T$	25 (298.15)	°C (K)	$k$	$1.7 \times 10^{-5}$	-
$l_{\max}$	10	mm	$p$	$1.42 \times 10^{-10}$	-
$C_{\text{Cu(I)}}^*$	$2.81 \times 10^{-7}$	mol/L	$D$	$1.2 \times 10^{-9}$	m <sup>2</sup> /sec
$C_{\text{Cu(II)}}^*$	$7.8737 \times 10^{-3}$	mol/L	$D_{\text{O}_2}$	$10^{-9}$	m <sup>2</sup> /sec
$C_{\text{O}_2}^*$	$2.01 \times 10^{-4}$	mol/L	$c$	0.022	1/mV
$C_{\text{Cl}}^*$	0.2	mol/L			

(a)

(b)

Table 3.4: Test parameters of the chloride model

Figure 3.3: Cu(I), Cu(II), and O<sub>2</sub> profiles obtained from the model. Note, that the scales for Cu(II) and O<sub>2</sub> are narrow, due to little variation in their concentrationsFigure 3.4: Relative profiles obtained from the model showing insignificant change in concentration of Cu(II) and O<sub>2</sub>

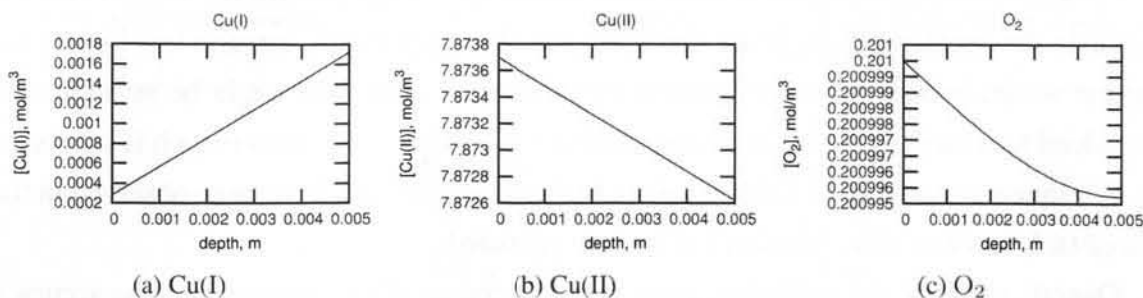


Figure 3.5: Concentration profiles for 5 mm capillary with calibrated values of reaction rates

rate of Cu(I) oxidation in contrast to that, reported in the literature for the stirred tank experiments (Nicol *et al.*, 2010), see Discussion section furtherbelow..

Using rate values found during calibration, the model has been validated against experimental data for 5 and 2 mm capillaries. The result of the validation is shown in table 3.6 on page 55. Although concentration of Cu(I) significantly differs from the experimental value, the deviation of the potential (the only value that has been really measured) stays with 1.1%, since its dependence on [Cu(I)] is logarithmic. Additionally, experimental results for the value of [Cu(I)] highly depend on the accuracy of measurements. Therefore, even with some variations in exact values, the model follows the general trend of the experiment. Profiles for the 5 mm capillary are shown in figure 3.5. The major difference between profiles 3.3 and 3.5 is a more linear form of Cu(I) and Cu(II) profiles due to lower value of the oxidation reaction rate coefficient  $k$ .

### 3.4 Discussion

Almost linear profiles obtained after model calibration suggest that there is very little influence of oxygen on the overall process. Therefore, the process is mainly governed by the rate of removal of the leaching product, Cu(I), from the mineral surface due to diffusion. Consequently, the longer the diffusion distance the slower is the process.

Low calibrated value of  $k$  results in the practical absence of Cu(I) oxidation. There is one possible scenario that might have mislead this conclusion: if the concentration of oxygen in the bulk solution was very low, then it would lead to extremely low values of  $k$ ,

which depends on the oxygen concentration, and to almost flat profiles for oxygen concentration in the capillary. Thus, if the concentration of oxygen in this scenario was higher, the process would be still governed by the removal of Cu(I), but there might be an additional considerable contribution to its removal, resulting in higher overall rates (which is supported by Velasquez-Yevenes *et al.* (2010b), since higher leaching rates have been observed in the stirred tank reactors if the solution had been oxygenated).

Overall, although the model has provided the estimate of the leaching rate, the accuracy of this estimate highly depends on the accuracy of measurements. Taking into account, that the derivation of concentrations from potential measurements is unstable, even slightly inaccurate measurement can lead to significantly different results. Thus, the estimate of leaching rate needs to be determined by actual concentration measurements (this is elaborated in the Discussions section 4.7 of the next chapter).

Parameter	Value	Unit
$\gamma$	$3.16 \times 10^{-5}$	-
$k$	$3.580 \times 10^{-7}$	l/sec
$p$	$8.778 \times 10^{-11}$	mol/(m <sup>2</sup> · sec)
$B$	$3.7166 \times 10^{-3}$	mol/m <sup>3</sup>
$E$	622.0	mV

Table 3.5: Chloride model calibration: final values of  $\gamma$  and parameters

Quantity	Experimental Value	Modelling Value	Relative Difference
For 10 mm capillary (used for model calibration)			
[Cu(I)]	$2.9 \times 10^{-3}$	$2.9 \times 10^{-3}$	0%
[Cu(II)]	7.8718	7.8716	0.0025%
$E$	622	622	0%
For 5 mm capillary			
[Cu(I)]	$1.82 \times 10^{-3}$	$1.72 \times 10^{-3}$	5.5%
[Cu(II)]	7.8726	7.8726	0%
$E$	634	635	0.16%
For 2 mm capillary			
[Cu(I)]	$1.1 \times 10^{-3}$	$0.83 \times 10^{-3}$	24.5%
[Cu(II)]	7.8731	7.8733	0.0025%
$E$	647	654	1.08%

Table 3.6: Chloride model validation: the comparison of the experimental values with modelling values



University of Cape Town

## Chapter 4

# General Modelling Platform Development

Although a relatively simple model, developed in the previous chapter, is capable of analysing experimental data, it is not suitable to model a heap environment. Since heap leaching is a dynamic process that involves advection (as an addition to diffusion) as a means of species transport, a more comprehensive model is required. The development of the agglomerate scale model is presented in this chapter.

As the word “model” has quite a broad meaning, it is important to specify the meaning of this word in different context:

**General model** denotes the set of equations and expressions that describes physical phenomena of the particular system. For example, a material point with mass  $m$  that moves under the force  $\mathbf{F}$  (and with some initial conditions) can be described by the Newton law of motion:

$$m\ddot{\mathbf{x}} = \mathbf{F}, \quad \mathbf{x}(0) = \mathbf{x}_0, \quad \dot{\mathbf{x}}(0) = \mathbf{v}_0, \quad (4.1)$$

which is the model for this physical system. A general model can be abstract if it is not completely defined (e.g. if initial conditions are omitted, or if source terms are not specified). The model (4.1) is an abstract model unless the form of the force  $\mathbf{F}$  is specified. If the model is set up completely but does not provide any means to solve its equations, we will call it theoretical model. In contrast, a practical model is the

model that provides an algorithmic tool (i.e. the tool that takes input data and after a finite time gives output result) to find the solution of its equations. Providing such a means usually implies the development of the numerical scheme and a computer programme that implements this scheme. However, there can be other ways which avoid development of a numerical scheme. For example, mechanical systems can be modelled by electrical circuits and the solution of equations can be measured as a current (or potential) in such circuits.

**Specific models** refer to particular parts of the physical processes. For example, a form of the force  $F$  in (4.1) is a specific model of the force field or a source term  $S$  in a diffusion equation (2.8) defines a specific reaction model. Specific models, required by general models to complete their formulation, usually derived from experimental data and are empirical or semi-empirical.

The aim of this chapter is to develop a practical general model of heap leaching that can be applied to a wide range of leach processes, i.e. allows incorporation of a wide range of specific reaction models. Additionally, the application of this general model to chloride leaching of chalcopyrite is demonstrated, thus extending the model developed in the previous chapter.

## 4.1 General description

The main focus of the research is on an agglomerate of particles and reaction-diffusion phenomena inside it. An agglomerate itself has a non-trivial structure to study. It can be seen consisting of:

**Ore particles** Each particle has a complex shape and mineralogy. Additionally, minerals are unevenly distributed over the particle surface.

**Solution with reagents** Some portion of the solution is virtually stagnant whereas there is a fraction that flows around particles. Some portion of the solution stays inside ore particles occupying the space in cracks. Reagents are unevenly distributed in the solution.

**Air** that occupies the rest of the space. Air either can be stagnant or can move depending on the operation.

As a result, the geometry and the internal structure of the particular agglomerate is highly complex. However, since the objective of the research project is to study a process in a typical agglomerate the following assumptions can be made:

- The agglomerate itself has a simple shape: cubical, spherical, cylindrical. Moreover, it can possess some symmetrical properties. For example, if the agglomerate has a cylindrical shape, the distributions of reagents and minerals are assumed to be cylindrically symmetrical, i.e. concentration of species can depend on the radius but not on the angle:

$$C_{\alpha} = C_{\alpha}(r), \quad \frac{\partial C_{\alpha}}{\partial \varphi} = 0,$$

where  $C_{\alpha}$  is a concentration of the species  $\alpha$ .

- Ore can be represented either by spherical particles (comprehensive approach) or by a semi-infinite "grey mass" (simplified approach, this approach, for example, has been used in the HeapSim, Ogbonna *et al.* (2005)). If ore is described in terms of ore particles, their size distribution can be involved for more sophisticated picture.
- Some parameters are uniformly distributed throughout the whole agglomerate, for example, temperature, initial mineral grade, etc.
- Moving and stagnant solution are separated. Since the flow inside agglomerate is obstructed, this part of solution is assumed to be stagnant, while the flow mainly takes place around the agglomerate. However, there might be no strict division into two phases, but instead, the flow rate can gradually decrease from the agglomerate boundary to the centre.

The above assumptions lead to a conceptual agglomerate with somewhat averaged properties and structure. Exact abstraction depends on the objective of research. For example, if the objective is to study competitive mineral leaching, the solution phase (outside ore particles) can be represented as single phase with uniform distribution of reagents. In contrast, if the

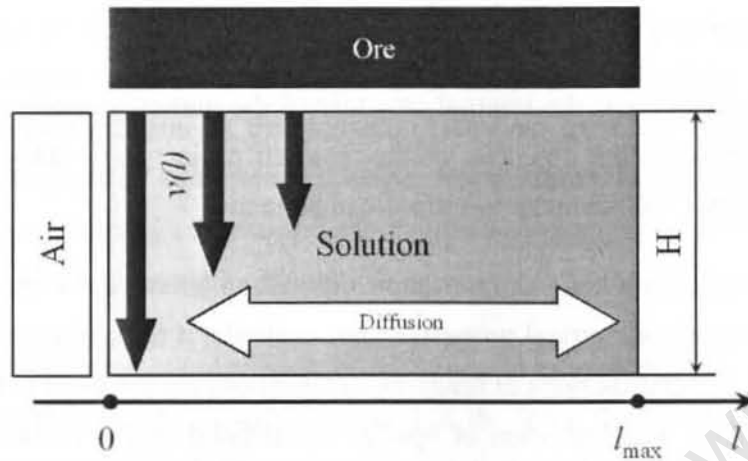


Figure 4.1: Schematic representation of the agglomerate of the height  $H$  and the depth  $l_{\max}$

objective is to find and examine effects due to transport phenomena in the solution phase, ore can be represented as a “grey mass” with uniform mineral distribution whereas the solution space has to have a complex structure. However, the model can be formulated in a way that it can shift the focus without large changes involved. Indeed, mathematically the model is a set of diffusion equations (see next section) regardless where the diffusion takes place.

A schematic representation of the agglomerate is shown in figure 4.1. The main focus on this figure is on the solution and species distribution in it. Thus, the ore is represented as some bulk non-moving material, which is in contact with the solution. Dissolved species can move across the agglomerate (in  $l$ -direction) due to diffusion, and can enter or leave it with advective flow which is defined by the velocity distribution  $v(l)$  perpendicular to  $l$ -direction. Solution can be in contact with air either throughout the whole volume or only at the specific part of the boundary of the solution domain. The latter imposes a mass transfer restriction on the migration of species (for example, oxygen) between liquid and gaseous phases, since different regions of the solution domain have different distances from the gas-liquid interface (figure 4.1). The former, in contrast, results in the less restricted migration of species between phases.

## 4.2 Mathematical description

### 4.2.1 Governing equation

As it was outlined before, the exact transport problem depends on the scale of the phenomena. The model, developed in this chapter, intends to simulate the agglomerate or the column (stack of agglomerates). As a result, it involves two transport phenomena: diffusion in the single agglomerate and advection, that transports species between agglomerates. It can be summarised by the unsteady-state advection-diffusion equation with a source term:

$$\frac{\partial C_\alpha}{\partial t} = \nabla \cdot \mathbf{D}_\alpha \nabla C_\alpha - \mathbf{v} \cdot \nabla C_\alpha + S_\alpha, \quad (4.2)$$

where  $C_\alpha$  is a concentration of species  $\alpha$ ,  $\mathbf{D}_\alpha$  is the diffusivity coefficient. Generally,  $\mathbf{D}_\alpha$  can be a second order tensor, i.e., considered as a linear operator which acts on the gradient  $\nabla C_\alpha$ , or a scalar value, and it may vary with position in space.  $S_\alpha$  represents the source term due to chemical reactions. To formulate the problem for a system with several species, equation (4.2) must be stated for each of them. The source term  $S_\alpha$  usually depends on the number of different species, thus, all equations must be solved simultaneously. Since the extensive use of the subscript  $\alpha$  can be distracting, it is omitted in some equations below, helping to keep focus on important ideas.

The first term on the right-hand-side (RHS) of equation (4.2) shows the effect of diffusion (with Fick's law applied). The exact form of the term depends on the geometry of the system (domain). For a rectangular system it is reduced to the second order derivative (assuming that the coefficient  $D$  is a scalar constant):

$$\nabla \cdot D \nabla C = D \nabla^2 C = D \left( \frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right).$$

If there is no dependency on spatial variables  $y$  and  $z$ , then this equation has a simplified 1D-form ( $x$  is replaced by  $l$ ):

$$\nabla \cdot D \nabla C = D \frac{\partial^2 C}{\partial l^2}.$$

If the system is cylindrical and cylindrically symmetrical (i.e. properties of the system do not depend on the angle, they can depend on the distance from the centre/surface and the vertical

position), then the term has the form as follows (assuming, that direction of  $l$  is opposite to that of radius, i.e.  $l = R_{\max} - R$ ):

$$D\nabla^2 C = D \left( \frac{\partial^2 C}{\partial l^2} - \frac{1}{R_{\max} - l} \frac{\partial C}{\partial l} + \frac{\partial^2 C}{\partial z^2} \right).$$

The spherically symmetrical system has a similar form for the diffusion term (but in this case 3D-domain is reduced into 1D one):

$$D\nabla^2 C = D \left( \frac{\partial^2 C}{\partial l^2} - \frac{2}{R_{\max} - l} \frac{\partial C}{\partial l} \right).$$

Both cylindrical and spherical systems represent the reduction in the availability of surface for the diffusion as  $l$  approaches its maximum value. It should be noted, that stated forms of the diffusion term for these systems are not always suitable for the numerical solution. The alternative approach is discussed in section 4.3.1.

The second term on RHS of (4.2) represents the effect of advection due to solution flow. A superficial velocity  $v$ , in general, can have arbitrary direction and can vary in the space. However, it is assumed that advection and diffusion phenomena occur in orthogonal directions, i.e. their effects are not directly coupled. For example, if the domain is rectangularly symmetric with spatial variables  $x$  (horizontal direction, denoted as before by  $l$ ) and  $z$  (vertical direction), the governing equation (4.2) has the form:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial l^2} - v(l, z) \frac{\partial C}{\partial z} + S. \quad (4.3)$$

Thus, while the value of the superficial velocity  $v$  can depend on the position in space, its direction is constant. A similar expression can be written for a cylindrical system (diffusion is assumed to be only in  $l$ -direction):

$$\frac{\partial C}{\partial t} = D \left( \frac{\partial^2 C}{\partial l^2} - \frac{1}{R_{\max} - l} \frac{\partial C}{\partial l} \right) - v \frac{\partial C}{\partial z} + S.$$

In contrast, this assumption is not directly applied to a spherical system since the advection term is going to destroy its symmetry. However, it can be incorporated if it is assumed that the solution flow occurs around the agglomerate, i.e.  $v = v_{\max}$  at the surface of the sphere ( $l = 0$ ) and  $v = 0$  at all other points ( $0 \leq l < l_{\max}$ ).

To summarise, the agglomerate model is assumed to be two-dimensional with the diffusion and advection taking place in orthogonal directions  $l$  and  $z$  respectively. In this case

coefficient  $D$  is a scalar value. Since the exact geometry of the system depends on application and the general form of equations does not depend on the geometry, it is more convenient to keep this general form in vector notation until the model is applied to a particular problem.

The form of the source term  $S_\alpha$  on RHS of the governing equation (4.2) depends on the choice of the reaction model. As a result, it can be defined only for each specific process. For the system with several reactions, it has the general form

$$S_\alpha = \sum_{\beta} \nu_{\alpha\beta} r_\beta, \quad (4.4)$$

where  $\nu = \|\nu_{\alpha\beta}\|$  is a stoichiometric matrix of reactions,  $r_\beta$  is the rate of reaction  $\beta$ . Therefore, the presence of source terms for several species systems, as it has been mentioned above, results in coupling of equations (4.2), written for each species.

### Non-moving species

Equation (4.2) is only applied to those species that can move. In contrast, if a species does not move, for example a mineral, its governing equation has a simplified form (however, it still can be considered as a special case of equation (4.2) with  $D = 0$  and  $v = 0$ ):

$$\frac{\partial C_\alpha}{\partial t} = S_\alpha. \quad (4.5)$$

### 4.2.2 Initial and boundary conditions

In order to complete the mathematical formulation of the problem, boundary and initial conditions must be added. Initial condition for the governing equation is the initial concentration of the reagent (vector  $x$  defines the position in the domain, it can refer to a 3D-vector, in the general case, or to a 2D-vector):

$$C_\alpha(t = 0, x) = C_\alpha^0(x). \quad (4.6)$$

Boundary conditions are more complex. Usually, boundary value problems are stated with the mixed conditions:

$$C_\alpha \Big|_{\partial\Omega_1} = C_\alpha^*, \quad (4.7)$$

$$-D \nabla C_\alpha \cdot n \Big|_{\partial\Omega_2} = N_\alpha, \quad (4.8)$$



where  $\partial\Omega$  is the surface boundary of the domain and  $\partial\Omega = \partial\Omega_1 \cup \partial\Omega_2$ , and vector  $\mathbf{n}$  is normal to the surface. For example, if the system involves advective flow, concentration at the entry surface may be stated. However, it is not always sensible to specify the concentration at the boundary without an advective flow. In this case Neumann boundary conditions are specified on the whole surface. For future application the surface is still split into two parts, as at the one part of the surface the flux is stated to be zero:

$$-D\nabla C_\alpha \cdot \mathbf{n} \Big|_{\partial\Omega_1} = 0, \quad (4.9)$$

$$-D\nabla C_\alpha \cdot \mathbf{n} \Big|_{\partial\Omega_2} = \mathcal{N}_\alpha. \quad (4.10)$$

The generalised boundary conditions for rectangular and cylindrical systems are defined as follows. At the mineral surface  $l_{\max}$  (or in the centre of the agglomerate, in this case  $R_{\max} = l_{\max}$ ) the flux of the species is specified:

$$-D \frac{\partial C_\alpha}{\partial l} \Big|_{l=l_{\max}} = \mathcal{N}_\alpha. \quad (4.11)$$

If the species participates in the mineral surface reaction,  $\mathcal{N}_\alpha$  is calculated on the base of the reaction rate, or it equals to zero otherwise (if  $l = l_{\max}$  corresponds to the centre of agglomerate,  $\mathcal{N}_\alpha = 0$  due to the symmetry of the system). As a result, boundary condition (4.11) is equivalent to (4.10).

At the liquid-air interface there are two possible cases. If the species cannot leave the domain at this boundary, its flux is stated to be zero (equivalent to (4.9)):

$$-D \frac{\partial C_\alpha}{\partial l} \Big|_{l=0} = 0. \quad (4.12)$$

However, if it can leave or enter the domain (as, for example, oxygen) the concentration at the boundary is specified (which is a particular case of (4.7)):

$$C_\alpha(t, l=0) = C_\alpha^*. \quad (4.13)$$

The surface concentration  $C_\alpha^*$  can be found, for example, by using appropriate thermodynamic correlations.

It is important to note, that if species do not leave or enter the domain at the liquid-air surface, i.e. they have (4.11) and (4.12) as boundary conditions, it is impossible to formulate

a correct steady-state problem, as this problem would have multiple solutions. In contrast, for the unsteady-state problem (4.2) it is possible to obtain a unique solution due to the presence of initial conditions (4.6).

### 4.2.3 Specific models

Although specific models depend on the particular heap leaching process they are designed to represent, it is still possible to define some general form of diffusion coefficient  $D_\alpha$  and rate terms  $r_\beta$  (see equation (4.4)).

Since the system temperature is one of the important parameters, both diffusion coefficient  $D$  and rate term  $r$  are stated to be a function of temperature and can be expressed by using Arrhenius terms:

$$D_\alpha = D_{\alpha \text{ ref}} \exp \left( -\frac{E_{a\alpha}}{R} \left( \frac{1}{T} - \frac{1}{T_{\alpha \text{ ref}}} \right) \right), \quad (4.14)$$

$$r_\beta = r_{\beta \text{ ref}} \exp \left( -\frac{E_{a\beta}}{R} \left( \frac{1}{T} - \frac{1}{T_{\beta \text{ ref}}} \right) \right), \quad (4.15)$$

where  $E_{a\alpha}$  and  $E_{a\beta}$  are activation energies of diffusion of species  $\alpha$  and reaction  $\beta$  respectively, and  $R$  is the universal gas constant. A typical value of activation energy  $E_\alpha$  for diffusion is about 15–25 kJ/mol (for example 19.2 kJ/mol has been reported for cupric ions, Moats *et al.*, 2000), which is lower than that of chemical reactions,  $E_\beta$ , resulting in complex (and often experimentally unpredictable) changes in the process as temperature changes.

Coefficient  $D_{\alpha \text{ ref}}$  usually is a constant, whereas a rate coefficient  $r_{\beta \text{ ref}}$  is determined by the choice of the reaction model and usually is a function of species concentrations (and maybe other factors).

## 4.3 Numerical scheme

The model definition (4.2) consists of partial differential equations with non-linear terms (source terms). The solution of this set of equations is performed in several steps.

First, equation (4.2), which can represent the stack of agglomerates (in rectangular and cylindrical coordinates), is split into a set of equations that describe the leaching process in a single agglomerate. This is performed by discretising equation (4.2) in the vertical

direction  $z$  (the direction of the advective flow) by finite differences method using backward derivatives (index  $\alpha$  is omitted):

$$\frac{\partial C_q}{\partial t} = \nabla \cdot \mathbf{D} \nabla C_q - v \frac{C_q - C_{q-1}}{H} + S_q, \quad (4.16)$$

where  $q$  is an agglomerate index and  $H$  is the height of the agglomerate. In this equation  $\nabla$  denotes the derivative in the direction orthogonal to  $z$ .

The next step is to discretise the space in the direction of diffusion, which is performed by applying the Bubnov-Galerkin scheme of FE methods.

### 4.3.1 Application of finite elements method

The result of application of the FE method is similar to (2.38) but it takes into account the presence of time derivative and advection term:

$$\mathbf{M} \frac{d\mathbf{C}_{\alpha,q}}{dt} + \mathbf{K}_{\alpha} \mathbf{C}_{\alpha,q} + \mathbf{F}_{\alpha,q} = 0, \quad (4.17)$$

where mass matrix  $\mathbf{M}$  is defined as follows:

$$M_{ij} = \int_{\Omega} \varphi_i \varphi_j d\sigma.$$

Matrix  $\mathbf{K}_{\alpha}$  incorporates diffusion and advection:

$$K_{\alpha,ij} = \int_{\Omega} \mathbf{D}_{\alpha} \nabla \varphi_j \cdot \nabla \varphi_i d\sigma + \int_{\Omega} \frac{v}{H} \varphi_j \varphi_i d\sigma.$$

For species with mixed boundary conditions (4.7) and (4.8) the last term has the same form as in (2.37):

$$F_{\alpha,qi} = - \int_{\Omega} S_{\alpha} \varphi_i d\sigma - \int_{\Omega} \frac{v}{H} C_{\alpha,q-1} \varphi_i d\sigma + \int_{\partial\Omega_2} \mathcal{N}_{\alpha} \varphi_i d\sigma + \int_{\Omega} \mathbf{D}_{\alpha} \nabla \overline{C}_{\alpha} \cdot \nabla \varphi_i d\sigma.$$

However, if Neumann boundary conditions apply, it leads to a slightly different system. In this case, the solution of the weak form of equation (4.2) belongs to Sobolev's space:

$$\mathcal{H}^1 = \left\{ f(\mathbf{x}) \in L_2(\Omega) \mid \int_{\Omega} (f^2 + (\nabla f)^2) d\sigma < \infty \right\},$$

which is similar to the space  $\mathcal{S}_0$  defined in equation (2.33) on page 31. Test functions are chosen from the same space:  $w(\mathbf{x}) \in \mathcal{H}^1$ . Once the domain is meshed and the discretised

sub-space  $\mathcal{H}^{1h}$  is constructed as a linear span of basis functions  $\varphi_i$  (see section 2.5.3), the projection of the solution into this sub-space has the form:

$$C_{\alpha,q}(t, \mathbf{x}) = \sum_j C_{\alpha,qj}(t) \varphi_j(\mathbf{x}).$$

Substitution of this expression into the weak form of equation (4.2) results in equation (4.17) where vector  $\mathbf{F}_\alpha$  has the form:

$$F_{\alpha,qi} = - \int_{\Omega} S_{\alpha} \varphi_i d\sigma - \int_{\Omega} \frac{v}{H} C_{\alpha,q-1} \varphi_i d\sigma + \int_{\partial\Omega_2} \mathcal{N}_{\alpha} \varphi_i d\sigma.$$

Before continuing discretisation of equation (4.17) further, it is sensible to obtain this equation for particular cases of rectangular, cylindrical and spherical systems.

#### Application to rectangular, cylindrical and spherical domains

The difference between systems comes from the different way of calculating the particle volume  $d\sigma$ . In rectangular coordinates it equals ( $\mathbf{x}$  is replaced by  $l$ )

$$d\sigma = dl.$$

While in cylindrical form (where  $R$  is a radius and  $l = R_{\max} - r$ ):

$$d\sigma = R dR = -(R_{\max} - l) dl.$$

And for the spherical system it equals to the following expression

$$d\sigma = R^2 dR = -(R_{\max} - l)^2 dl.$$

Thus, the resulting ODEs for rectangular and cylindrical systems look as follows (respectively):

$$\begin{aligned} \sum_j \frac{dC_j}{dt} \int_0^{l_{\max}} \varphi_j \varphi_i dl + \sum_j C_j \int_0^{l_{\max}} D \varphi_j' \varphi_i' dl - \sum_j C_j \int_0^{l_{\max}} \frac{v(l)}{H} \varphi_j \varphi_i dl + \\ \int_0^{l_{\max}} \frac{v(l)}{H} C_{\text{inlet}} \varphi_i dl - \int_0^{l_{\max}} r \varphi_i dl + \mathcal{N} \varphi_i \Big|_{l=l_{\max}} = 0, \quad (4.18) \end{aligned}$$

$$\begin{aligned}
& \sum_j \frac{dC_j}{dt} \int_0^{l_{\max}} \varphi_j \varphi_i (R_{\max} - l) dl + \sum_j C_j \int_0^{l_{\max}} D \varphi_j' \varphi_i' (R_{\max} - l) dl - \\
& \sum_j C_j \int_0^{l_{\max}} \frac{v(l)}{H} \varphi_j \varphi_i (R_{\max} - l) dl + \int_0^{l_{\max}} \frac{v(l)}{H} C_{\text{inlet}} \varphi_i (R_{\max} - l) dl - \\
& \int_0^{l_{\max}} r \varphi_i (R_{\max} - l) dl + \mathcal{N} \varphi_i (R_{\max} - l) \Big|_{l=l_{\max}} = 0, \quad (4.19)
\end{aligned}$$

where  $C_{\text{inlet}}$  denotes the concentration at the previous agglomerate (since index  $q$  is omitted, otherwise  $C_{\text{inlet}} = C_{q-1}$  and  $C = C_q$ ) and  $\varphi_i' = d\varphi_i/dl$ .

The equation for the spherical system, as has been mentioned before, does not involve the advection:

$$\begin{aligned}
& \sum_j \frac{dC_j}{dt} \int_0^{l_{\max}} \varphi_j \varphi_i (R_{\max} - l)^2 dl + \sum_j C_j \int_0^{l_{\max}} D \varphi_j' \varphi_i' (R_{\max} - l)^2 dl - \\
& \int_0^{l_{\max}} r \varphi_i (R_{\max} - l)^2 dl + \mathcal{N} \varphi_i (R_{\max} - l)^2 \Big|_{l=l_{\max}} = 0. \quad (4.20)
\end{aligned}$$

Thus, it can be used to describe the diffusion inside ore particles.

### 4.3.2 ODE solution

Ordinary differential equations (4.17) are solved by applying the implicit Euler method to insure the absolute stability (see section 2.5.4):

$$\left( \frac{1}{\Delta t} \mathbf{M} + \mathbf{K}_\alpha \right) \mathbf{C}_\alpha^{k+1} = \frac{1}{\Delta t} \mathbf{M} \mathbf{C}_\alpha^k - \mathbf{F}_\alpha^{k+1}, \quad (4.21)$$

where  $\Delta t$  is a step of the time discretisation. The set of non-linear equations (4.21) is solved by the direct application of the contraction mapping method (see section 2.5.5):

$$\left( \frac{1}{\Delta t} \mathbf{M} + \mathbf{K}_\alpha \right) \mathbf{C}_{\alpha(n+1)}^{k+1} = \frac{1}{\Delta t} \mathbf{M} \mathbf{C}_\alpha^k - \mathbf{F}_{\alpha(n)}^{k+1} = \mathbf{B}_{\alpha(n)}^{k+1}, \quad (4.22)$$

where  $n$  is the index of approximation. This set of linear equations (4.22) is solved by the bi-conjugate gradient method (BiCG).

## 4.4 Model implementation

### 4.4.1 Object structure of the programme

The programme has adopted the object-oriented programming (OOP) style which has not been widely used for numerical computer modelling until recent time. The OOP approach provides number of advantages:

- Re-use of the existing code.
- Adaptation of the existing code for the particular problem.
- Since components interact via their interfaces, the internal structure of each component is independent from others.
- Reduction in the number of global variables and parameters that need to be passed to functions.

The programme is represented by the following objects (or classes in terms of C++):

**Heat** Class `Heat` handles the implementation of FEM. The functionality of the class relies on the external library DEAL.II (Bangerth *et al.*, 2007). In the initial form, this class has been used for a heat conduction problem in the DEAL.II documentation and that is from where it has its name.

**Species** Class `Species` keeps the information related to individual species, such as diffusivity, activation energy, concentration, etc.

**SourceTerm and BCSourceTerm** These classes are used to calculate rates of volumetric and surface reactions (`SourceTerm` and `BCSourceTerm` respectively).

**IOData** This class encapsulates the functionality for data input and results output.

**Model** This is the governing class, that puts together the functionality of other classes. Additionally, the non-linear solver algorithms is implemented in this class.

Some utility objects are omitted in the description above due to their technical role in the programme. The full listing of the programme can be found in Appendix A.

```
while current time is less than overall time do
    Increase current time by the time step  $\Delta t$ ;
    Update time step information;
    Solve non-linear system (4.21);
    Output data if necessary;
end
```

**Algorithm 4.1:** Main algorithm

#### 4.4.2 Description of programme algorithms

The main algorithm of the programme runs the simulation over all time steps, finishing when the overall time of simulation is reached (algorithm 4.1). It is implemented in the function `solve()` of the class `Model`. This algorithm uses `one_time_step_solve()` function to solve the non-linear equation (4.21) at each time step.

Function `one_time_step_solve()` is schematically shown in algorithm 4.2. This function implements the contraction mapping method for the column simulation model. It iterates over agglomerates, starting from the top. For each agglomerate it solves the non-linear equation (4.21) by constructing the series of solutions of linear systems (4.22). The solution converges, when the relative difference (in the combined maximum- and  $L_2$ -norms) between approximated solutions (`rel_diff`) became smaller than predefined value  $\varepsilon$ . Additionally, the method uses the variable `steps_num` to protect the programme from infinite looping if the solution does not converge. If this happens, the programme prints an error message with the available information and terminates. The construction and solution of the linear system is delegated to the function `next_approx()` of the class `Heat`.

Overall, the programme and the main algorithm structures enable one to adopt this programme for different applications since the change of types of reagents and kinetics parameters can be done easily. However, this will still require considerable changes in the source code.



```

for each agglomerate do
    { number of iterations counter }
    steps_num  $\leftarrow$  0;
    repeat
        { check if there are too many iterations }
        if steps_num > MAX_STEPS then
            { Solution does not converge within certain amount
              of steps }
            Report an error and finish the programme;
        end
        steps_num  $\leftarrow$  steps_num + 1;
        Solve linear system (4.22) for each species;
        Calculate rel_diff =  $\max_{X=[A], [B], [C]} \frac{\|\Delta X\|}{\|X\|}$ ;
    until rel_diff <  $\varepsilon$ ;
    if the current agglomerate is not a bottom one then
        Set inlet concentrations for each species in the below agglomerate to be equal
        to the concentrations of the species in the current agglomerate;
    end
end

```

Algorithm 4.2: Method of contraction mapping

## 4.5 Test of the model: simple case study

The model has been applied to a simple case study in order to test its stability and convergence. The variety of physical and chemical conditions (such as reaction rates, temperature, diffusivity coefficients, etc.) were applied as well as different numerical conditions.

### 4.5.1 Test case description

The system with three reagents A, B and C is considered. They can react as follows (volumetric reactions):







The rates of these reactions are described by the the expressions:

$$r_1 = k_1(T) \frac{[A]^2}{([B] + 1)}$$

$$r_2 = k_2(T) \frac{[B]^2}{([C] + 1)}$$

Since these expressions are non-linear, the non-linear solver can be tested on them. Additionally, two rates can make the problem stiff enabling to check the efficiency of the time discretization method.

The agglomerate is assumed to be rectangular and all system parameters are independent from the spatial variables  $l$  and  $z$ . Thus, equation (4.3) is used to describe the system.

To complete the description of the system, diffusion equations are stated for each species:

$$\frac{\partial C_A}{\partial t} = D_A(T) \frac{\partial^2 C_A}{\partial l^2} + v(l) \frac{C_{\text{inlet},A} - C_A}{H} - r_1, \quad (4.23)$$

$$\frac{\partial C_B}{\partial t} = D_B(T) \frac{\partial^2 C_B}{\partial l^2} + v(l) \frac{C_{\text{inlet},B} - C_B}{H} + r_1 - r_2, \quad (4.24)$$

$$\frac{\partial C_C}{\partial t} = D_C(T) \frac{\partial^2 C_C}{\partial l^2} + v(l) \frac{C_{\text{inlet},C} - C_C}{H} + r_2, \quad (4.25)$$

where velocity profile has the form  $v(l) = v_{\max} \exp(-\lambda l)$  to insure that it reaches maximum at  $l = 0$  and decreases to 0 as  $l$  increases. At the boundaries of the domain ( $l = 0$  and  $l = l_{\max}$ ), species do not enter and do not leave the system, i.e. conditions for each species are

$$-D \frac{\partial C}{\partial l} \Big|_{\text{boundary}} = 0$$

This system provides facilities to test it against the change of diffusivity coefficients  $D(T)$ , reaction rates coefficients  $k(T)$ , superficial velocity  $v(l)$ , domain height  $H$  and domain length  $l_{\max}$ .

To demonstrate a general model output, the simulation has been performed with parameter values listed in tables 4.1, 4.2 and 4.3 on page 74. Sample results are shown in figure 4.2:

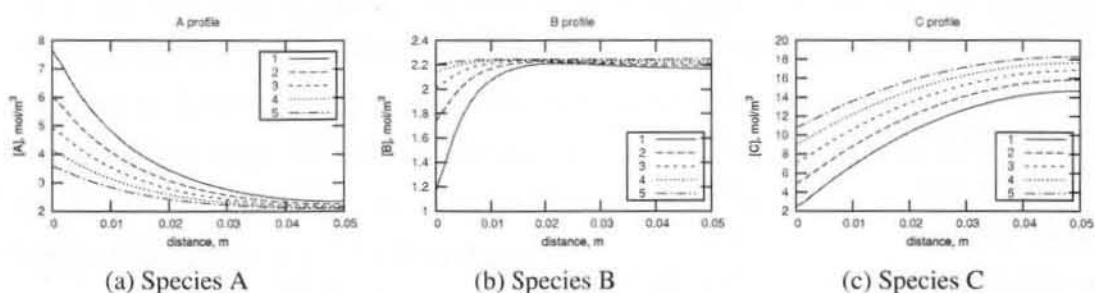


Figure 4.2: A sample diffusion profile of species A, B and C over the stack of agglomerates ("1" denoting the top agglomerate) after 10 days

- Species A is the only species that gets introduced into the system ("reagent") from the top agglomerate (with the highest amount entering at  $l = 0$  where superficial velocity  $v(l)$  has a maximum). In the system itself A is consumed due to the chemical reaction. Therefore, the profile shows the maximum concentration at  $l = 0$  that gradually decreases as  $l$  approaches  $l_{\max}$ . As species A is consumed at each agglomerate, its overall concentration falls from the top agglomerate to bottom one.
- Since species B is both produced and consumed in the system ("intermediate product"), its profile largely depends on the difference between reaction rates. The drop in the concentration of B at  $l = 0$  is due to advection since species B does not enter the domain with the flow.
- Species C is the main product of the system. Thus, its profile is somewhat opposite of the profile of A. Since it does not enter the system with the advective flow, but only leaves with it, the lowest concentration of C is at  $l = 0$ . The concentration is accumulated with the advective move from top to bottom.

#### 4.5.2 Tests on different grids

In order to achieve approximation to the real solution, the numerical solution of equations (4.23), (4.24) and (4.25) needs to be independent from the grid (space discretization step). The independence from the grid indicates the stability of the numerical scheme.

Parameter	Value	Unit
Total time	10	days
Initial time step	$4 \times 10^{-6}$	days
Output time step	0.9	days
Domain size	0.05	m
Max superficial velocity	$5 \times 10^{-5}$	m/sec
Velocity factor $\lambda$	2500	–

Table 4.1: Test model parameters

Reaction	Rate constant $k_{\text{ref}}$
$A \rightarrow B$	$2 \times 10^{-6}$
$B \rightarrow C$	$1.67 \times 10^{-5}$

Table 4.2: Reaction rate parameters

Species	Initial Concentration	Inlet Concentration	Diffusivity
A	10 mol/m <sup>3</sup>	10 mol/m <sup>3</sup>	$10^{-9}$ m <sup>2</sup> /sec
B	10 mol/m <sup>3</sup>	0 mol/m <sup>3</sup>	$10^{-9}$ m <sup>2</sup> /sec
C	5 mol/m <sup>3</sup>	0 mol/m <sup>3</sup>	$10^{-9}$ m <sup>2</sup> /sec

Table 4.3: Species parameters

Mesh numbers	Species		
	A	B	C
32 and 64	0.1%	0.16%	0.17%
64 and 128	0.09%	0.04%	0.05%
128 and 256	0.04%	0.02%	0.025%

Table 4.4: Differences ( $l_2$ -errors) between solutions on different grids for species A, B and C

The model was tested with the parameters listed in tables 4.1, 4.2 and 4.3 and on a grid of 32, 64, 128 and 256 elements. Since solutions for different grids lie in different spaces, they all have been projected on to the space with the least number of elements (32 in this case), i.e. all other points were discarded. After this they all become vectors with 32 components and can be directly compared. To check the convergence, the  $l_2$ -error has been calculated between projected vectors from different grids.

The result of the  $l_2$ -error calculation for each species is shown in the table 4.4. As it can be seen, all differences decrease as the grid number becomes larger. Additionally, the difference for each transition is not larger than 0.2%. Thus, the result of the present model simulation is fairly independent of grid number, indicating the stability of the numerical scheme (the solution of a mesh-dependent problem changes dramatically as the mesh gets refined or coarsened, for example, the solution of a diffusion-advection problem starts oscillating if the mesh is coarsened beyond a particular point, Chizhonkov (2006)).

### 4.5.3 Test with different time steps

Since the time step changes during computation depending on how fast convergence is being achieved, the effect of the initial time step is not obvious. The main goal of this test is to check how significant this effect can be.

Tests have been performed with parameters listed in tables 4.1, 4.2 and 4.3 with 32 elements in the mesh. The initial time step has been chosen as  $4 \times 10^{-6}$  days (0.3456 sec) and has been gradually increased up-to  $3.6 \times 10^{-4}$  days (31.104 sec). The overall  $l_2$ -error between the control solution (with the smallest initial time step) and other solutions does not exceed 0.01% for each species. Therefore, model results are essentially independent of the choice of the initial time step in the present case (the application of the forward Euler method would result in the solution dependence on the time-step, since if the time-step is too large, the discretised solution can start oscillating and growing, whereas the solution of the original continuous problem is monotonous and decreasing, see section 2.5.4).

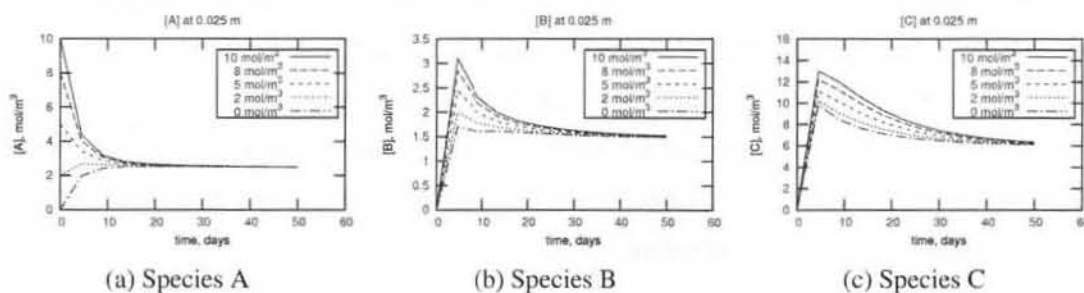


Figure 4.3: Change of species concentrations with time depending on the initial concentration of species A (the sharp peak in the profiles of species B and C is due to coarse recording time step)

#### 4.5.4 Effect of initial conditions

Since the case study model is a dynamic model (there is a time derivative in equations (4.23), (4.24), (4.25)), the solution does depend on initial conditions. However, neither boundary conditions nor source terms depend on time explicitly. Furthermore, this model is non-linear and involves advection and, therefore, has essential (Dirichlet) boundary condition at the top of the two-dimensional domain. Therefore, as time tends to infinity, the solution approaches a steady state which should not depend on initial conditions (in contrast, a linear problem  $u_t - a\nabla^2 u = b$ , with only Neumann BC specified, significantly depends on the initial condition  $u_0$ , since if it is changed to  $u_0 + u^*$  the solution will shift  $u(t) + u^*$ ).

The model has been tested on five different initial conditions of the species A (the only initial reagent in the system, since other species are either intermediate or final products): 10, 8, 5, 2, and 0 mol/m<sup>3</sup>. Results shown in figure 4.3 suggest that after 50 days the difference between models with diverse initial conditions disappears.

#### 4.5.5 Effect of reaction rates coefficients

As was indicated before, a diffusion problem combined with the reaction phenomena can result in a stiff system. Stiffness appears in systems where there is a large difference between rates of variable changes. For the test case system this can appear if there is a large difference between reaction rates.

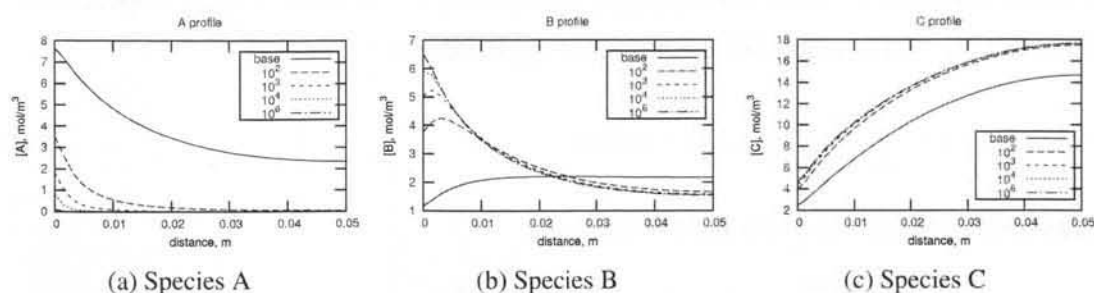


Figure 4.4: Change of profiles with the change of the reaction rate constant  $k_1$  from the base value to  $10^2$ ,  $10^3$ ,  $10^4$  and  $10^6$  faster.

The reaction rate of the second reaction has been kept as before,  $1.67 \times 10^{-5}$  1/sec, while the rate of the first reaction has been drastically increased from  $2 \times 10^{-6}$  1/sec to 2 1/sec. Since the impact of the first reaction was increased dramatically, this resulted in the stiff problem. Although backward Euler algorithm is not absolutely stable for stiff problems, in this case it was still feasible to stabilise it by keeping low initial time step.

The result of the test is shown in figure 4.4. The most dramatic changes have occurred with the profile of species B: because of much higher production rate it has formed a maximum near  $l = 0$  where is the highest concentration of species A.

Overall, although algorithm convergence required smaller initial time step when the ratio of rate coefficients exceeded  $10^4$  due to rapid initial changes, the model still showed stability and continuous change of the result with the change of the reaction rate coefficient  $k_2$ .

#### 4.5.6 Mass balance

The test model has been checked for consistency in terms of the mass balance. According to stoichiometry of the system, the overall amount of all species initially present and entering through the inlet over the time must be equal to the overall final residual amount and total amount left through the outlet over the time.

The residual amount of species (say, A) at time  $t$  can be expressed as an integral (discarding horizontal surface area, which remains constant):

$$\text{RES}(C_A, t) = \sum_{\text{agglomerates}} \int_0^{l_{\max}} C_A(l, t) H dl. \quad (4.26)$$

Then initial and final amounts can be found substituting  $t = 0$  and  $t = t_{\text{total}}$  respectively.

The total inlet amount comes from the top agglomerate:

$$\text{IN}(C_A, t) = \int_0^{l_{\text{max}}} C_{\text{inlet},A}(l, t) \cdot v(l) dl \quad (4.27)$$

whereas total outlet leaves from the bottom one:

$$\text{OUT}(C_A, t) = \int_0^{l_{\text{max}}} C_A^{\text{bottom}}(l, t) \cdot v(l) dl \quad (4.28)$$

Then mass balance at time  $t_{\text{total}}$  can be expressed as follows:

$$\sum_{i=A,B,C} \left( \text{RES}(C_i, 0) + \int_0^{t_{\text{total}}} \text{IN}(C_i, t) dt \right) = \sum_{i=A,B,C} \left( \text{RES}(C_i, t_{\text{total}}) + \int_0^{t_{\text{total}}} \text{OUT}(C_i, t) dt \right) \quad (4.29)$$

In the programme integration over the spatial variable has been conducted using Gaussian quadrature rule (order 2), while integration over the time has been approximated by the trapezoidal method. The programme was run on parameters listed in tables 4.1 and 4.3 and on the domain uniformly meshed with 32 elements. The result of simulation shows that the overall relative error in mass balance over the total time of simulation (300 days) is less than 0.06%. The low value of this error, introduced by both problem approximation and limited computer precision, proves the accuracy of the numerical scheme. The change of the mass balance error with time is shown in figure 4.5. Initially, the error changes quite chaotically, since there is a rapid change in species concentration and accumulated error can be cancelled out with the next time step. Between 10 and 60 days species concentration changes gradually, and the error stays within 0.01%. Between 70 and 150 days the error grows quite rapidly, but does not exceed 0.03%. After 150 days the error growth declines. Overall, after 300 days the error stays within 0.06% thus well controlled.

#### 4.5.7 Case study conclusions

The case study has demonstrated stability and consistency of the formulated model over quite large range of parameter values. However, some test results (for example, dependence on



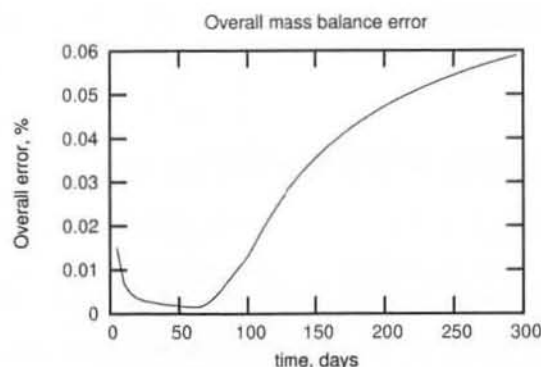


Figure 4.5: The dependence of mass balance error on time

$l_{\max}$ ) can be meaningless for this simplified model, since it does not account for the presence of the mineral in the system. Thus, these tests are performed on a more sophisticated and practical case.

## 4.6 Application to chloride leaching

This section demonstrates the application of the generic model to the chloride leaching of chalcopyrite. This application aims to extend the model, developed in chapter 3 for the experimental analysis, to enable it to analyse the results of the column tests.

Agglomerate model for the chloride leaching, as well as the simple model from the previous chapter, consists of only one dissolved species, Cu(I). Additionally, chalcopyrite mineral,  $\text{CuFeS}_2$ , that initially is uniformly distributed through the agglomerate, has been added to investigate the progress of the mineral depletion.

Since the mineral is spread through the whole domain, i.e. it is not situated just at the boundary, the source term due to mineral leaching has been moved from the boundary condition (3.14) into the equation itself (here  $C = [\text{Cu(I)}]$ ):

$$\frac{\partial C}{\partial t} = D \nabla^2 C - v \frac{\partial C}{\partial z} - 4k(C - C^*) + 4\beta r_{\text{cpy}}, \quad (4.30)$$

where  $\beta$  is a coefficient that is responsible for the transformation of the surface reaction into volumetric one.



Mineral depletion is described via equation:

$$\frac{\partial X}{\partial t} = \alpha \beta r_{\text{cpy}}, \quad (4.31)$$

where  $X$  is a mineral conversion, defined as follows:

$$[\text{CuFeS}_2] = [\text{CuFeS}_2]_0(1 - X),$$

coefficient  $1/4$  is due to reaction stoichiometry (see (3.1)) and  $\alpha$  recasts the dimension of  $\beta r_{\text{cpy}}$  (for the detailed derivation of  $\alpha$  and  $\beta$  see next section). Source term  $r_{\text{cpy}}$  is defined similarly to (3.17), but with added dependence on the mineral conversion:

$$r_{\text{cpy}} = p(T)g(E)(1 - X)^\varphi, \quad (4.32)$$

where  $\varphi$  is an empirical parameter, that determines the depletion of the surface available for leaching.

Initial conditions are stated trivially:  $C(t = 0) = C_{\text{init}}$ ,  $X(t = 0) = 0$ . With regards to boundary conditions, concentration at the top of the agglomerate is specified  $C(z = 0) = C_{\text{inlet}}$ , whereas there is no flux through the side boundaries:  $\mathcal{N}(l = 0, l_{\text{max}}) = 0$ .

#### 4.6.1 Definition of transformation coefficients

To complete the system, coefficients  $\alpha$  and  $\beta$  need to be defined. Their role is to transform the dimension of the mineral leaching source term  $r_{\text{cpy}}$  to match that of the respective equation:

$$\begin{aligned} r_{\text{cpy}} & [=] \frac{\text{mol}}{\text{m}_{\text{cpy}}^2 \cdot \text{sec}}, \\ \beta r_{\text{cpy}} & [=] \frac{\text{mol}}{\text{m}_w^3 \cdot \text{sec}}, \\ \alpha \beta r_{\text{cpy}} & [=] \frac{1}{\text{sec}}. \end{aligned}$$

Equation (4.31) can be written in the form:

$$\frac{\partial C_{\text{cpy}}}{\partial t} = -\frac{\varepsilon_w}{\varepsilon_s} \beta r_{\text{cpy}},$$

where  $C_{\text{cpy}}$  is a concentration of chalcopyrite, defined as  $\text{mol}/\text{m}_{\text{ore}}^3$ . Thus, in terms of mineral conversion,  $X$ , it has the form:

$$\frac{\partial X}{\partial t} = \frac{1}{C_{\text{cpy}0}} \frac{\varepsilon_w}{\varepsilon_s} \beta r_{\text{cpy}}.$$

And coefficient  $\alpha$  is defined as follows:

$$\alpha = \frac{1}{C_{\text{cpy } 0}} \frac{\varepsilon_w}{\varepsilon_s} = \frac{M_{\text{cpy}}}{g_0 \rho_{\text{ore}}} \frac{\varepsilon_w}{\varepsilon_s} = \frac{M_{\text{cpy}}}{\bar{g}_0 \rho_{\text{cpy}}} \frac{\varepsilon_w}{\varepsilon_s},$$

where  $M_{\text{cpy}}$  is molar mass of chalcopryrite,  $\rho$  is density,  $g_0$  is mass mineral grade ( $\text{kg}_{\text{cpy}}/\text{kg}_{\text{ore}}$ ) and  $\bar{g}_0$  is volumetric mineral grade ( $\text{m}^3_{\text{cpy}}/\text{m}^3_{\text{ore}}$ ).

Definition of  $\beta$  is largely based on the assumption that ore in the agglomerate consists of spherical particles with some distribution of the radius  $r$ .

Since  $r_{\text{cpy}}$  defines the rate of the surface reaction, to transform it to refer to volumetric reaction (per volume of solution), it needs to be multiplied by the following coefficient:

$$\beta = \frac{S_{\text{cpy}}}{V_w} = \frac{\bar{g}_0 S_{\text{ore}}}{\varepsilon_w V},$$

where  $S_{\text{cpy}}$  is the total surface of chalcopryrite in the agglomerate available for leaching and  $V$  is a total volume of the agglomerate.

The value of  $S_{\text{ore}}$  can be determined from the ore particle distribution. If  $p(r)$  is a density of this distribution, then

$$p(r)dr = \frac{n(r)}{N}dr$$

defines the portion of particles with radius between  $r$  and  $r + dr$ , where  $n(r)$  is a the number of particles with radius  $r$  and  $N$  is the total number of particles in the agglomerate. Using this density function, the total surface is defined as follows:

$$S_{\text{ore}} = \int_0^{R_{\text{max}}} 4\pi r^2 p(r) N dr$$

The total number of particles can be found from their volumes:

$$N = \frac{\varepsilon_s V}{\int_0^{R_{\text{max}}} \frac{4}{3}\pi r^3 p(r) dr}$$

Finally, the expression for  $S_{\text{ore}}$  looks as follows:

$$S_{\text{ore}} = \frac{1}{3} \varepsilon_s V \frac{\int_0^{R_{\text{max}}} r^2 p(r) dr}{\int_0^{R_{\text{max}}} r^3 p(r) dr} = \frac{1}{3} \varepsilon_s V \frac{\int_0^{R_{\text{max}}} r^2 df(r)}{\int_0^{R_{\text{max}}} r^3 df(r)}, \quad (4.33)$$

where  $f(r)$  is a portion of particles with the radius less than  $r$ :

$$f(r) = \int_0^r p(r) dr$$

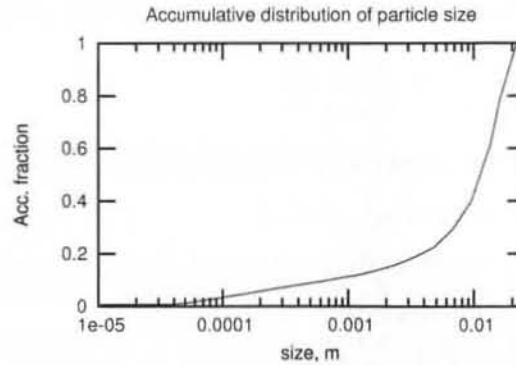


Figure 4.6: Accumulative distribution of particle size,  $f(r)$

The distribution of particles after ore crushing is usually defined by the tabulated values of  $f(r)$ . If this information is available, then integrals in (4.33) can be approximated with the following expression:

$$S_{\text{ore}} \approx \frac{1}{3} \varepsilon_s V \frac{\sum_{i=1}^I (r_{i-1}^2 + r_i^2) (f(r_i) - f(r_{i-1}))}{\sum_{i=1}^I (r_{i-1}^3 + r_i^3) (f(r_i) - f(r_{i-1}))}$$

#### 4.6.2 Model results

To test the model, the programme has been run on the parameters listed in tables 4.5, 4.6, 4.7. While some values are well known (such as copper ion diffusivity  $D$ ), others are speculative. Additionally, values of the rate constants  $p$  and  $k$  have been chosen from the calibration of the model from the previous chapter (see section 3.3). The value of  $S_{\text{ore}}$  has been calculated based on some typical distribution of ore particles after crushing shown in figure 4.6 (J. Petersen, personal communication, 2010).

Parameter	Value	Unit	Comment
$v_{\max}$	$3 \times 10^{-3}$	m/sec	$v(l) = v_{\max} \exp(-2.5 \times 10^3 l)$
$l_{\max}$	$10^{-2}$	m	0.5 cm
Domain height	0.5	m	
Total agglomerate volume	$1.57 \times 10^{-4}$	$\text{m}^3$	$V = \pi l_{\max}^2 H$
Number of agglomerates	1	—	
$\varepsilon_s$	0.5	—	
$\varepsilon_w$	0.2	—	
Number of elements	16	—	
Temperature	25	$^{\circ}\text{C}$	298.15 K
Total time	300	day	
Initial time step	$10^{-6}$	day	0.09 sec

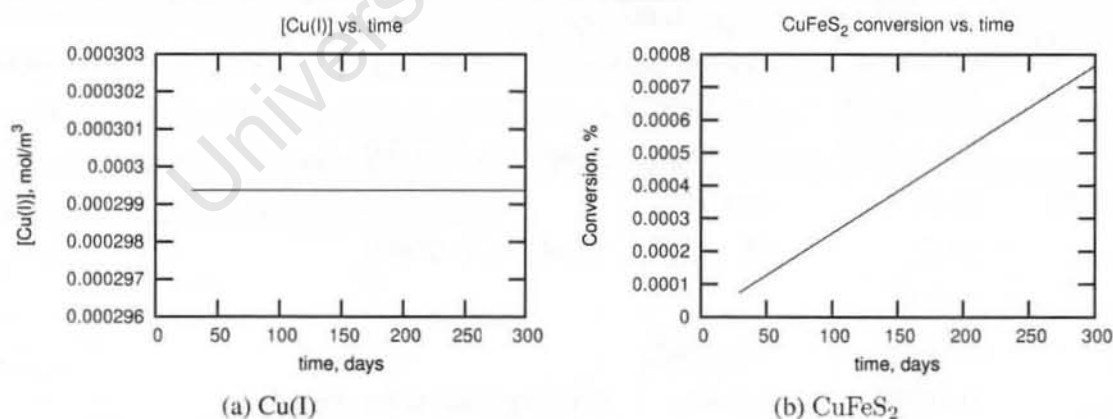
Table 4.5: General model parameters

Parameter	Value	Unit	Comment
$C^*$	$2.807 \times 10^{-4}$	$\text{mol}/\text{m}^3$	
$D$	$1.2 \times 10^{-9}$	$\text{m}^2/\text{sec}$	Moats <i>et al.</i> (2000)
$[\text{Cu}^{2+}]$	6.18	$\text{mol}/\text{m}^3$	
$E_{a, \text{Cu}^+}$	19.2	kJ	Moats <i>et al.</i> (2000)
$T_{\text{ref}, \text{Cu}^+}$	298.15	K	
$\bar{g}_0$	0.001	$\text{m}_{\text{cpy}}^3/\text{m}_{\text{ore}}^3$	
$M_{\text{cpy}}$	0.18354	kg/mol	Chalcopyrite molar mass
$\rho_{\text{cpy}}$	4190	$\text{kg}/\text{m}^3$	Chalcopyrite density

Table 4.6: Species related parameters

Parameter	Value	Unit	Comment
$\varphi$	2/3	—	
$p$	$8.78 \times 10^{-11}$	$\text{mol}/\text{m}^2 \cdot \text{sec}$	See table 3.5
$E_{a,p}$	72	kJ	Activation energy for the leaching reaction
$T_{\text{ref},p}$	298.15	K	
$k$	$3.58 \times 10^{-7}$	$\text{mol}/\text{m}^3 \cdot \text{sec}$	See table 3.5
$E_{a,k}$	70	kJ	Activation energy for the oxidation reaction
$T_{\text{ref},k}$	298.15	K	
$S_{\text{ore}}$	$1.450 \times 10^{-2}$	$\text{m}^2$	See equation (4.33)
$\alpha$	$1.75 \times 10^{-3}$	$\text{m}_w^3/\text{mol}_{\text{cpy}}$	
$\beta$	4.62	$1/\text{m}_w$	

Table 4.7: Reaction rates related parameters

Figure 4.7: Change of concentration of Cu(I) and the conversion of CuFeS<sub>2</sub> with time at  $l = 2.5 \text{ mm}$

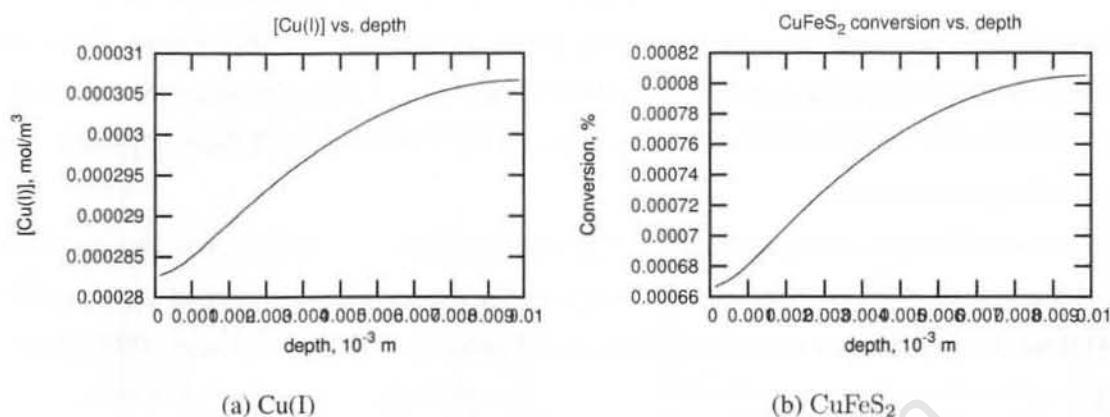


Figure 4.8: Variation of the concentration of Cu(I) and the conversion of CuFeS<sub>2</sub> after 300 days. Note, the small scale showing very small variation within the agglomerate

Results of this model can be summarised as follows:

- Due to low value of the rate coefficient  $p$ , the leaching rate is extremely slow, even at elevated temperature: after 300 days mineral conversion reached only 0.0008% (figure 4.7).
- The concentration of Cu(I) seems do not change over the time (figure 4.7) due to low reaction rate, resulting in a pseudo-steady-state process.
- There is almost no variation neither in concentration of Cu(I) nor in mineral conversion over the space since the reaction rates are slower than diffusion (see figure 4.8).

These almost flat profiles are due to low leaching reaction rate (i.e. low value of  $p$ ). Similar mineral conversion value can be obtained with a rough calculation. If it is assumed that all ore particles are spherical with the same radius  $r = 5$  mm, which is the mean value of the distribution shown in figure 4.6), then the volume of one particle can be estimated approximately as  $5 \times 10^{-7} \text{ m}^3$ . The total volume of the agglomerate is  $\pi l_{max}^2 H = 1.57 \times 10^{-4} \text{ m}^3$ . Thus, the total volume of the ore is  $\varepsilon_s V_t \approx 0.79 \times 10^{-4} \text{ m}^3$ . The total number of ore particles can be calculated as  $N = V_s / V_{\text{ore particle}} = 150$ . The total surface area of the ore particles is  $4\pi r^2 N = 0.047 \text{ m}^2$ . If the volumetric grade of chalcopyrite is 0.1% the total area of chalcopyrite accessible for leaching can be estimated as  $4.7 \times 10^{-5} \text{ m}^2$ . If the leaching

reaction rate is assumed to be  $9 \times 10^{-11} \text{ mol}/(\text{m}^2 \cdot \text{sec})$  then the total dissolution rate can be estimated as  $2 \times 10^{-7} \text{ mol/day}$  or, converting it into percentage,  $2 \times 10^{-5}\%/ \text{day}$ . Thus, after 300 days this will yield the conversion of 0.006%, which is, although higher than the model prediction, is still considerably lower than rates shown in column tests (see Appendix A for details of the calculation).

The model results change dramatically if the leaching rate coefficient is increased: figures 4.9 and 4.10 show the profiles obtained from the model with the value of  $p$  chosen to be 1000 times higher. In this case the overall chalcopryrite conversion increases 1000 times as well.

## 4.7 Discussions and further research

The results of the model simulation demonstrated quite a slow leaching process, which would not be economically feasible. However, some preliminary column studies (Petersen, 2008) had shown faster overall leaching rate (conversion rate of about 0.1-0.2% a day) than that predicted by the model. This discrepancy suggests that

- Either rate parameters  $p$  and  $k$  do not have correct values, or
- Current understanding of the reaction mechanism (which is summarised in equations (3.1) and (3.2)) is not complete.

In either case, systematic experimental study is required.

Those studies should focus on the interaction of leaching reactions and mass transfer effects, trying to keep the number of parameters to a minimum. Taking the design of the pore-diffusion experiment of Basson (2010) as a basis, the following modifications should be made to obtain more accurate results:

- Potential measurements do not provide accurate estimation of the leaching rate, since the deduction of the rate relies on the inverse of the Nernst equation (2.5):

$$\frac{[\text{Cu}^+]}{[\text{Cu}^{2+}]} = \exp\left(\frac{F}{RT}(E^0 - E)\right),$$

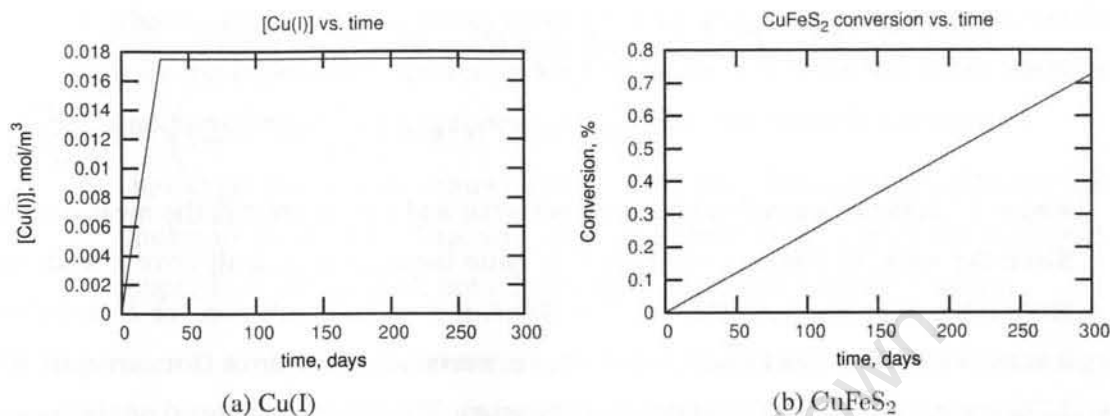


Figure 4.9: Change of concentration of Cu(I) and the conversion of CuFeS<sub>2</sub> with time at  $l = 2.5$  mm for the artificially increased leaching rate

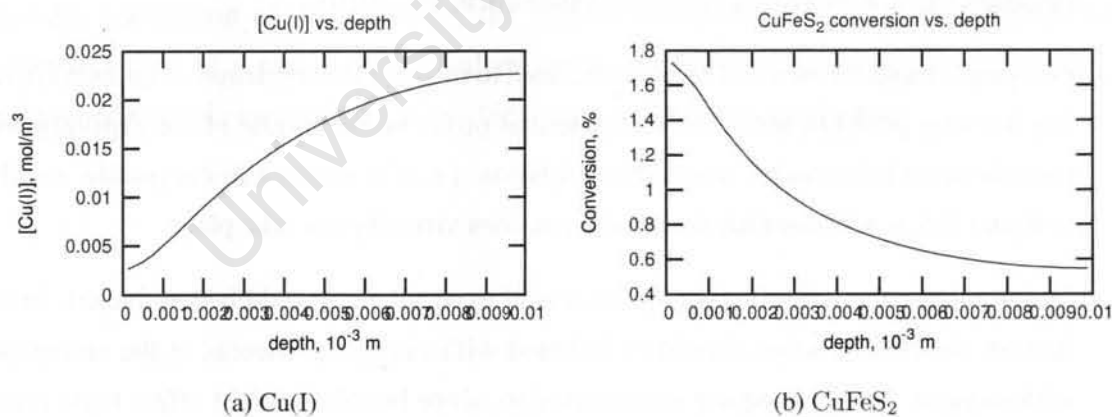


Figure 4.10: Variation of the concentration of Cu(I) and the conversion of CuFeS<sub>2</sub> after 300 days for the artificially increased leaching rate (similarly to figure fig:abc-init, the sharp peak in Cu(I) concentration is due to coars recording time step, 30 days)



which makes the calculation unstable. Indeed, a small inaccuracy in the measurement introduces a significant error in the concentration value, that affects the rate:

$$\exp\left(\frac{F}{RT}(E^0 - E^*)\right) = \exp\left(\frac{F}{RT}(E^0 - E) + \frac{F}{RT}\varepsilon\right) = \exp(F/(RT)\varepsilon) \exp\left(\frac{F}{RT}(E^0 - E)\right) = \delta \frac{[\text{Cu}^+]}{[\text{Cu}^{2+}]},$$

where  $E^*$  is the estimated value of the potential and  $\varepsilon$  is an error in the measurement. Since the value of Faraday constant  $F$  is quite large, even a small error  $\varepsilon$  leads to a large value of  $\delta$ . For example, for  $T = 300$  K if  $\varepsilon = 0.002$  mV (i.e. for  $E = 0.6$  mV it is merely 0.33% error) then  $\delta = 1.08$ , i.e. introducing 8% error (for  $\varepsilon = 0.01$  mV, 1.7% error,  $\delta = 1.47$ , i.e. introduces 47% error). Thus, rates calculated on the base of potential measurements can include a significant error and, hence, it is preferable to measure concentrations directly.

Since there is a high background concentration of copper and the change in the overall concentration is quite small, it is not possible to achieve accurate measurements of changes of copper concentration. However, if the system starts iron-free, the overall leaching rate can be well estimated by the measurement of iron concentration (see equation (3.1), J. Petersen, personal communication, 2010).

- Potential measurements still needs to be used to monitor the oxidation of copper(I) (3.2). For the long pore (10 mm) an extra potential probe in the middle of the capillary will provide more information about this oxidation, i.e. if it shows a linear profile, similar to figure 3.5, it will confirm that oxidation does virtually not take place.
- To investigate the role of oxygen, two sets of experiments should be conducted. In the first set the bulk solution should be bubbled with nitrogen, whereas in the second set with oxygen. It might require some redesign, since bubbling might affect mass transfer. It has been proposed to use a layer of sand instead of capillary as it would disrupt any advection in the solution that fills pores between sand grains (J. Petersen, personal communication, 2010). There are three possible outcomes of these experiments:
  1. No difference between the two sets of experiments. Thus, oxygen is really unimportant. Although this outcome is possible it is highly unlikely, since it had been

indicated that in stirred tank reactor oxygenated systems showed faster leaching rates (Velasquez-Yevenes *et al.*, 2010a).

2. The oxygenated reactor shows faster leaching rates, and species profiles, obtained from the experiment, are non-linear (see figure 3.3). This will prove the current understanding of the oxygen role, as a reagent that controls potential.
  3. The oxygenated reactor shows faster leaching rates, but species profiles are linear, similar to figure 3.5. This will show that there is no significant oxidation of copper(I) in the solution, but oxygen plays some other role in leaching.
- Experiments at different temperatures with various pore lengths will show how significant the mass transfer effect is under various conditions. Since diffusion has a lower activation energy in comparison with reaction rates, its effect at lower temperatures can be insignificant, but will become more apparent as temperature grows. The effect of diffusion on the leaching rate at higher temperatures had already been indicated from the analysis of column tests data (Petersen, 2008). This study can isolate any other phenomena that can affect leaching rate at higher temperature and reconfirm mass transfer constraints.

After the completion of those studies, the pore-diffusion model can be re-calibrated with respect to values of  $p$  and  $k$ . These values can be used in the agglomerate-scale model to obtain more realistic estimations of leaching rates in column experiments.

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## Chapter 5

### Conclusions

The current work aimed to investigate possible mass transfer constraints on the chloride leaching of chalcopyrite at two different scales using modelling as a research tool.

The first question that has been answered was

What effect limits the leaching rate at the micro-scale?

The analysis has shown significant accumulation of cuprous ions near the mineral surface at longer pores, while concentrations of other species have changed insignificantly. Furthermore, this accumulation in longer pores resulted in slower leaching rates. Thus, it has been concluded that it is slow removal of cuprous ions (the product) from the reaction zone that limits the overall process.

To make sure that model results reflect the experimental data correctly, the model was systematically calibrated and validated. As an outcome of the calibration process, the leach rate coefficients have been obtained. The low value of the cuprous oxidation reaction rate, established from the calibration, suggested that oxygen in this case does not play a significant role in the removal of cuprous ions.

In parallel, a generic agglomerate-scale model was developed. This model aims to simulate column experiments and provide insight into the phenomena inside columns. Mathematically, it has extended the pore-diffusion model, used to analyse experimental data, by incorporating additional effects such as advection, non-linear source terms and time dependence. This extension required the development of more comprehensive numerical methods (introducing finite elements methods) and programme platform (using C++ programming

language, DEAL.II library for finite elements). These large changes required extensive tests of the new programme to make sure that the numerical scheme is stable and the programme is free of internal errors. The stability of the model was achieved by the application of a finite elements method to a cylindrically symmetrical domain (see section 4.3.1, as a direct application of FD can lead to unstable numerical scheme) and by the use of an implicit method for time integration. Sensitivity tests performed on the model have shown that the model results are independent from the mesh size, initial time step value and the choice of initial conditions (since the system is conservative). Overall, the programme offers an open platform that is robust, extendable and adaptable to various heap leach processes.

The model was applied to chloride leaching of chalcopyrite using the parameters derived from the initial analysis and using a realistic low-grade chalcopyrite ore. The results have shown that the leaching process with the given parameters would be extremely slow (0.01% of chalcopyrite converted after 300 days) and this has been confirmed with an order-of-magnitude calculation based on the initial data. However, preliminary column experiments have shown significantly higher rates of leaching at 0.1-0.2% a day (Petersen, 2008). This difference between the model results and the experimental data indicates that the process requires further research at the micro-scale. A series of experiments has been proposed to obtain more accurate estimates of the leach reaction rates. The results of these experiments then can be used to re-calibrate the model. It is recommended this be pursued in a further project.

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# Appendix A

## Spreadsheets and Source Code Listings

The CD, supplied with the thesis, consists of the following files/folders:

**ClAnalysis.ods** is a spreadsheet (Open Document format) that represents the linear model developed in section 3.1.

**ClExperiment** consists of files that implements pore-diffusion model (see section 3.2). Source files (in Scala programming language) are in the subfolder `cltestopt`:

**Solver.scala** implements the contraction mapping method to solve algebraic equations.

**Optimize.scala** implements the simulated annealing method to find the minimum of a function of two variables.

**Model.scala** holds model related functions, such as the function representing the Nernst equation, leaching rate function and etc.

**OptimumFinder.scala** holds two functions: one that represents the functional to be minimised,  $\gamma$  (see equation (3.24)), and another one that starts the optimisation process on the model.

**ClTest.scala** is responsible for input and output. It consists of only one function, `main`, that starts the process of the model calibration and prints the output.

Subfolder `classes` consists of the results of compilation the source files. Additionally, the script `run.sh` (for BASH shell) starts the model simulation. The compilation

and the run of the programme requires Scala Environment 6.8 (the installer is in the file `scala-2.8.0.RC2-installer.jar`) and Java Development Kit 1.6 to be installed.

**abc\_test** and **ClAgglomerate** consist of files for the case-study and the chloride models respectively. Since these programmes share common source base, their source-files are described together:

**solver\_bcg.h** implements BiCG method to solve the set of sparse linear equations.

**heat.h** and **heat.cc** implement the application of the Bubnov-Galerkin method to the diffusion-reaction problem. Initially, this application has been designed for the heat conduction problem and then has been adopted for the diffusion problem.

**species.h** and **species.cc** hold the information about a particular species such as a concentration distribution, diffusivity coefficient, etc.

**source\_term\_function.h** and **source\_term\_function.cc** represent volumetric reaction rates and source terms.

**init\_val.h** holds function to populate initial values for concentrations.

**model.h** and **model.cc** represent the model. Time integration, contraction mapping method, mass balance check are implemented in these files.

**bc.h** (only for the chloride model) represent essential boundary conditions. The functionality of this file is not used at the moment, but the presence of this file is essential for the **heat.cc** to compile correctly since the latter is designed in a generic way to be able to deal with species that have essential boundary conditions.

The rest of the files have an utility role, such as keeping constants (**info.h**), implementing input-output (**iodata.h** and **iodata.cc**) and starting the programme (**main.cc**).

The functionality of these two programmes relies on the external library, DEAL.II (the source code is packed in the archive `deal.II-6.2.1.tar.gz`), which is an open

source library distributed under the Q Public Licence (QPL, the text of the licence is included in the `LICENCE` file).

The compilation of the programmes is performed in several steps:

1. DEAL.II should be compiled first (requires g++ compiler of version 4.2 or newer).
2. The source files of the programmes should be compiled into object files. This compilation might require (depending on the system settings) the explicit pointing to include-directories of the DEAL.II library.
3. Object files should be linked with the DEAL.II shared or static library files (only `base`, `deal_II_1d` and `lac` libraries are required).

The compilation under Linux is straight-forward. However, the compilation under Windows requires some preparation steps:

1. Cygwin environment should be installed with the stable version of the compiler `gcc` and `g++`.
2. Since Cygwin is shipped with an older version of `g++`, a new version (4.2 or newer) should be compiled from source codes and installed under Cygwin.

**RateEstimate.ods** provides the estimation of the leaching rate using simple, although rough, calculations.